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1 **Effect of temperature cycle on mechanical properties of**  
2 **methane hydrate-bearing sediment**  
3

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15 **Abstract**

16 In this study, methane hydrate-bearing sand (MHBS) was created in the laboratory following  
17 two methods in order to obtain two types of gas hydrate morphology in sandy sediment. The  
18 hydrate morphology in the sediment was assessed by measuring the compressional wave  
19 velocity combined with models to predict the wave velocities of the sediment containing gas  
20 hydrates. The mechanical properties of the MHBS were investigated by triaxial compression  
21 tests. The results obtained by the compressional wave velocity show that after saturating the  
22 MHBS sediment (created by the excess gas method) with water, the methane hydrates are partly  
23 or completely converted from grain contacts to pore spaces depending on the hydrate saturation  
24 (ranging from 0 to 50%). A subsequent temperature cycle completes this conversion process  
25 for high hydrate saturation. The results obtained with the triaxial compression tests show higher  
26 shear strength, a higher secant Young's modulus, and a higher dilation angle at higher hydrate  
27 saturation. In addition, the effects of hydrate saturation on the mechanical properties of the  
28 MHBS obtained by the two procedures (with and without the thermal cycle) are similar at low  
29 hydrate saturation. The effect of gas hydrate morphologies can only be detected in the case  
30 where the conversion (and/or redistribution) of gas hydrates from grain contacts to pore spaces  
31 is not complete (at high hydrate saturation).

32

33 **Key words:** *methane hydrate, sand, mechanical behavior, pore habit, rock physics model.*

34

## 35 **1. Introduction**

36 Methane hydrates, being solid ice-like compounds of methane and water, form naturally at high  
37 pressure and low temperature. Due to the growing energy demand, natural gas hydrates  
38 (primarily methane hydrates) are being considered as an alternative energy source (Collett et  
39 al., 2009). However, methane hydrate dissociation during borehole drilling and the production  
40 process (with heat or depressurization methods) may reduce the strength of the hydrate-bearing  
41 sediments and cause failure (Nixon and Grozic, 2007). In addition, slope instability and wide-  
42 scale gas venting are the two most important geo-hazards associated with methane hydrate  
43 dissociation problems on the sea floor (Collett et al., 2014; Sultan et al., 2004). For these  
44 purposes, various studies have been performed to investigate the mechanical behavior of  
45 methane hydrate-bearing sediments.

46

47 In natural sediments, methane hydrates exist in the form of nodules/chunks, lenses/veins or  
48 pore-filling depending on the characteristic particle size and effective stress (Dai et al., 2012;  
49 Konno et al., 2015; Masui et al., 2008). After Collett et al. (2009), the majority (90%) of  
50 methane hydrates is said to be found in fine-grained sediments in dispersed forms, but hydrate  
51 saturation is typically low. On the contrary, the larger pore size and relatively high permeability  
52 of sandy sediments facilitate methane hydrate formation as pore-filling with high hydrate  
53 saturation. For this reason, methane hydrate-bearing sand (MHBS) is the actual target for  
54 potential gas hydrate exploration within the scope of future gas production.

55

56 In spite of the various numerical models (Jiang et al., 2014; Pinkert & Grozic, 2014; Sánchez  
57 et al., 2017; Uchida et al., 2012), few experimental works exist on the investigation of the  
58 mechanical behavior of MHBS. Among the existing experimental studies, few works focus on

59 intact samples (Masui et al., 2008; Winters et al., 2007; Yoneda et al., 2015, 2017). Almost all  
60 the experimental works are concerned with laboratory tests on synthetic samples because of the  
61 challenges involved with getting cored intact methane hydrate-bearing sediment samples. With  
62 recent advances in core pressure technology (pressure-core analysis and transfer system -  
63 PCATs), the temperature and pressure of samples can be maintained within hydrate stability so  
64 that mechanical tests on almost undisturbed samples can be done. However, improvements are  
65 still needed to increase the successful pressuring core proportion and to efficiently extrude  
66 samples into the PCAT's triaxial cell (Priest et al., 2015; Yamamoto, 2015; Yoneda et al., 2017,  
67 2015).

68

69 The hydrate morphology in sediments is usually assessed by comparing the measured seismic  
70 velocities and those calculated via models (Dvorkin and Nur, 1998, 1996; Helgerud et al.,  
71 1999). In the model proposed by Helgerud et al. (1999), four pore-scale hydrate distributions  
72 are considered. This model is based on the fact that seismic velocities depend on the bulk elastic  
73 moduli of the system, which are controlled by the grain-scale arrangements of the hydrates and  
74 the sediment. For a given hydrate saturation, the hydrates floating in the pore fluid (pore-filling  
75 morphology) increase the seismic velocity by increasing the modulus of the pore fluid. This has  
76 the smallest impact on the host sediment's elastic properties. On the contrary, hydrates forming  
77 only at the grain contacts and acting as cement (contact cement morphology) have the greatest  
78 impact on the elastic properties of sediments and increase the seismic velocity by locking  
79 individual grains together. The hydrates forming part of the sediment frame (load-bearing  
80 morphology) are simply considered as a second mineral in the quartz sand pack, while the  
81 hydrates surrounding and cementing the sediment grains (grain-coating morphology) are held  
82 together by the effective pressure. Their presence dramatically increases the granular contact  
83 stiffness by locking the grains in place.

84

85 The relationship between compressional velocity and hydrate saturation is usually used to  
86 assess the hydrate pore habit. In sandy samples, the gas hydrates can be created at the contacts  
87 between the sand grains in gas-saturated media by various methods: (i) ice seeding (Priest et  
88 al., 2005), (ii) excess gas (partial water saturation) without water saturation (Grozić and  
89 Ghiassian, 2010; Hyodo et al., 2013a; Waite et al., 2004; Winters et al., 2007; Zhang et al.,  
90 2012), (iii) or excess gas (partial water saturation) followed by water saturation (Hyodo et al.,  
91 2013a; Miyazaki et al., 2011b). Dissolving the gas is considered as a good method for  
92 reproducing natural methane gas hydrate formation in marine sediments, but this method is  
93 time-consuming especially at high hydrate saturation due to the low solubility of methane gas  
94 in water (Spangenberg et al., 2005). Priest et al. (2009) proposed the water excess method, but  
95 methane hydrates formed heterogeneously inside their sample (Kneafsey et al., 2010). Choi et  
96 al. (2014) proposed a non-cementing methane hydrate-forming method by combining the  
97 excess gas method with saline water injection at restricted conditions and a temperature cycle.  
98 However, in the work of Choi et al. (2014), mechanical tests were not performed to investigate  
99 the effect of the hydrate pore habit (hydrate morphology) on the mechanical properties of the  
100 sediment.

101

102 Without measuring the acoustic properties, Ebinuma et al. (2005) and Masui et al. (2005)  
103 formed methane hydrates by the excess gas and ice-seeding methods, determined the  
104 mechanical properties of MHBS (by triaxial compression tests), and suggested that pore-filling  
105 hydrates could be created by the ice-seeding method.

106

107 In the present work, gas hydrates were created in sandy samples following two methods. The

108 first one corresponds to the creation of methane hydrates in a partial water saturation sample  
109 followed by a water saturation phase prior to the mechanical testing. The second one was similar  
110 to the first one, but was completed with a temperature cycle (as suggested by Choi et al., 2014).  
111 That would lead to two types of hydrate pore habits. The measurement of the compressional  
112 wave velocity and triaxial compression tests were used to assess the grain-scale hydrate  
113 distribution in sediments. Tests were performed with various hydrate saturations (0 - 50%) to  
114 investigate the effect of this parameter on the mechanical properties of the MHBS. It should be  
115 noted that gas hydrate-bearing sandy sediments in nature could have higher hydrate saturation  
116 (up to 80%). In addition, other researchers (Yoneda et al., 2017; Yun et al., 2007) mentioned  
117 that the mechanical properties of hydrate-bearing sediments would change exponentially with  
118 hydrate saturation in the range over 40~50%. However, the hydrate pore habit would be more  
119 complex in these cases and would need more extensive work.

## 120 **2. Experimental method**

### 121 **2.1. Materials**

122 The soil used in this study was Fontainebleau silica sand (NE34). Its mechanical properties are  
123 generally well-documented (Dupla et al., 2007). It consists of poor-graded sub-rounded grains  
124 having diameters ranging from 100 to 300 microns (the particle size distribution curve shown  
125 in Figure 1 was obtained by laser diffraction analysis). Tap water was used in the tests. The  
126 standard purity of the methane gas used here was 99.995%.

### 127 **2.2. Experimental setup**

128 Figure 2 presents a schematic view of the temperature-controlled high-pressure triaxial  
129 apparatus. The sample (1) was covered with a neoprene membrane, 50 mm in diameter and 100  
130 mm in height. A displacement sensor (2) was used to monitor the radial strain of the sample,  
131 serving to calculate the volumetric strain during consolidation. For the mechanical loading, the

132 confining pressure was applied to the fluid (silicone oil) inside the cell by a volume/pressure  
133 controller (3); the deviator stress, applied via the piston (4), was measured by the force  
134 transducer (5) installed above the piston. It should be noted that the friction between the cell  
135 and the piston (corresponding to approximately 200 kPa of deviator stress) can be measured  
136 when the piston is moved down without touching the top baseplate (10). Methane gas was  
137 injected via the bottom inlet (6) by a pressure controller, which was connected to a gas  
138 flowmeter. The top and bottom pore pressure transducers were connected by a T-valve (7). The  
139 pore water pressure was controlled by a volume/pressure controller (8). For the temperature  
140 control, the cell was immersed in a temperature-controlled bath connected with a cryostat. A  
141 thermocouple (9) was placed close to the sample to measure the temperature inside the cell. For  
142 the measurements of the compressional wave velocities, two ultrasonic sensors were installed  
143 on the top and bottom baseplates (10) and connected to a wave generator.

### 144 **2.3. Test procedure**

145 MHBS samples were prepared by the following procedure:

- 146 - Step 1: Moist sand (having a known moisture content) was compacted by tamping in  
147 layers to obtain a void ratio of 0.63 inside the neoprene membrane prior to the assembly  
148 of the experimental setup, as shown in Figure 2.
- 149 - Step 2: The sample was consolidated at drained conditions (state 1 in Figure 3). The  
150 confining pressure was increased to 25 MPa then decreased to 10 MPa. As the maximal  
151 value of confining pressure during the subsequent test was 22 MPa, the consolidation  
152 step would ensure that the soil remained in the elastic domain during the whole test.
- 153 - Step 3: To create methane hydrates inside the sample, the temperature of the bath was  
154 decreased to 3 – 4°C and vacuum was applied to eliminate the pore air in the sample.  
155 Afterward, methane gas was injected at 7 MPa during the whole methane hydrate  
156 formation period (state 2 in Figure 3). The methane hydrate formation in gas-saturated



157 media was considered complete when the methane gas flow rate became negligible (<  
158 0.1 ml/min). At the end of this step, methane hydrate saturation can be estimated from  
159 the initial water saturation (an increase in volume of 10% was considered due to the  
160 water-hydrate shift). It should be noted that 7 MPa of gas pressure is much higher than  
161 the value required to create gas hydrates at a temperature of 3 - 4°C (see state 2 in Figure  
162 3). In addition, preliminary studies have shown that the ultrasonic sensors could  
163 correctly measure the compressional wave velocities only in the case of an effective  
164 stress higher than 1 MPa (Ebinuma et al., 2008; Rydzy and Batzle, 2010). For this  
165 reason, a confining pressure of 10 MPa (corresponding to an effective stress of 3 MPa)  
166 was chosen.

167 - Step 4: To saturate the MHBS sample, the T-valve (7) was opened to the atmosphere  
168 for a short period (about 10 s) to let the excess gas (initially under a pressure of 7 MPa)  
169 escape from the sample (i.e., the pore pressure decreased to zero) and then this valve  
170 was connected to the volume/pressure controller (8) to inject water with a pressure of 7  
171 MPa. This procedure allowed the excess gas to be replaced in the sample by water and  
172 minimized the disturbance of methane hydrates that already existed inside the sample  
173 by saturating the sample without circulating water.

174 After the above steps had been completed, two different procedures were used:

175 - For procedure A, a drained triaxial compression test was performed directly after  
176 verifying the good saturation of the sample at 22 MPa of confining pressure and 19 MPa  
177 of pore pressure by mean of Skempton's coefficient.

178 - For procedure B, a temperature cycle was performed beforehand to modify the gas  
179 hydrate distribution at the grain scale. For this purpose, the pore pressure and the  
180 confining pressure were first decreased from 7 MPa to 4 MPa and from 10 MPa to 7  
181 MPa, respectively. All the drainage valves were then closed and the temperature of the

182 cell was increased to higher than 20°C. That corresponds to a heating of the sample  
183 under undrained conditions in order to progressively dissociate the existing gas  
184 hydrates. During this phase, the pore pressure (measured by the pressure transducer)  
185 increased progressively because of the heating. A Labview program was used to  
186 automatically control the confining pressure (via the volume/pressure controller (3)) in  
187 order to maintain the mean effective stress (confining pressure minus pore pressure) at  
188 3 MPa. When the pore pressure reached 19 MPa and the confining pressure reached 22  
189 MPa, the T valve (7) was opened for connection with the volume/pressure controller  
190 (8). During this step, the pressure in the volume/pressure controller (8) was maintained  
191 at 19 MPa and the volume of water expelled during this heating period was monitored.  
192 The gas hydrates were supposed to be totally dissociated when the volume of expelled  
193 water had stabilized. After this dissociation phase, gas hydrates were then reformed in  
194 the sample by decreasing the temperature of the cell to 3-4°C, while maintaining the  
195 pore pressure at 19 MPa (and the confining pressure at 22 MPa). This phase induced an  
196 injection of water from the volume/pressure controller (8) to the sample. The hydrate  
197 reformation phase was considered to be finished when the volume of injected water had  
198 stabilized. After the gas hydrate reformation, a drained triaxial compression test was  
199 performed under an effective confining pressure of 3 MPa as in the case of tests using  
200 procedure A.

201 In the pore pressure vs temperature plot (Figure 3), procedure A follows the path (1)-(2)-(3),  
202 while procedure B follows the path (1)-(2)-(4)-(3). The axial strain rate for all the triaxial  
203 compression tests was fixed at 0.1%/min to ensure the drainage conditions. The final methane  
204 hydrate saturation was determined by measuring the volume of methane gas dissociated at the  
205 end of the triaxial compression tests.

## 206           **2.4. Test program**

207   The test program is shown in Table 1. Four tests were performed with procedure A (A1 to A4)  
208   and four tests were performed with procedure B (B1 to B4). The moisture content was imposed  
209   prior to the sample preparation. The water saturation was calculated from the imposed moisture  
210   content, the void ratio obtained after the sample preparation ( $e = 0.63$ ), and the particle density  
211   ( $\rho_s = 2.65 \text{ Mg/m}^3$ ). In addition, a reference test was performed, which consisted of compacting  
212   dry sand down to a void ratio of 0.63 in the triaxial cell. After the consolidation step (confining  
213   pressure was increased to 25 MPa and then decreased to 10 MPa), the sample was saturated  
214   with water at a confining pressure of 10 MPa and a water pressure of 7 MPa. Finally, a triaxial  
215   compression phase at 22 MPa of confining pressure and 19 MPa of pore pressure was applied  
216   as with the other samples. The hydrate saturations shown in Table 1 were determined at the  
217   end of the tests from the volume of methane gas dissociated from the sample.

## 218           **3. Experimental results**

### 219           **3.1. Isotropic consolidation**

220   Figure 4 and Figure 5 show the void ratio ( $e$ ) and the compressional wave velocity ( $V_p$ ) versus  
221   mean effective stress ( $p'$ ), respectively, during the consolidation step for all tests (Step 2). The  
222   points of the unloading phase correspond to the decrease in confining pressure from 25 MPa to  
223   10 MPa followed by the injection of methane gas at 7 MPa. The compression curves ( $e - p'$   
224   plot) show a decrease in void ratio from 0.63 to 0.58-0.59 during the loading path (up to  $p' =$   
225   25 MPa). During the unloading path, the void ratio increases to 0.60 – 0.61 (when  $p' = 3 \text{ MPa}$ ).  
226   The effect of the moisture content on the compression behavior is not clear; the small  
227   discrepancy in the results can be then related to the good repeatability of the experimental  
228   procedure.

229

230 Unlike the compression curves ( $e - p'$  plot), where hysteresis can be observed during the  
231 loading/unloading paths, the relationship between  $V_p$  and  $p'$  shows reversible behavior (Figure  
232 5). Actually, during the loading path when  $p'$  increases from 1.6 MPa to 25 MPa (the ultrasonic  
233 sensors used in this study could not measure  $V_p$  lower than 700 m/s),  $V_p$  increases from 750 –  
234 850 m/s to 1400 – 1500 m/s. During the unloading path when  $p'$  is decreased to 3 MPa,  $V_p$   
235 decreases to 900 – 1000 m/s. The results obtained in the consolidation step confirm that  $V_p$  is  
236 strongly dependent on the mean effective stress (Zimmer, 2004), whereas it is independent of  
237 the void ratio and moisture content. For instance, when  $p'$  equals 3 MPa, the void ratio at the  
238 loading path is higher than that at the unloading path, but the  $V_p$  obtained by the two paths are  
239 similar. This can be explained by the small volumetric strain (3%). In addition, the results  
240 shown in the  $V_p - p'$  plot for all the samples (having various moisture contents) are similar.  
241 Actually, the matric suction in sand (few kPa, after Feia *et al.*, 2016) is much smaller than the  
242 mean effective stress. All the subsequent measurements of  $V_p$  were performed at the same mean  
243 effective pressure (3 MPa). The discrepancy on the  $V_p$  (around  $\pm 50$  m/s) can be attributed to the  
244 repeatability of the experimental procedure.

### 245 **3.2. Methane hydrate formation**

246 Figure 6 plots  $V_p$  versus elapsed time during the methane hydrate formation (Step 3) in gas-  
247 saturated media (the confining pressure was maintained at 10 MPa and the gas pressure at 7  
248 MPa). At the beginning,  $V_p$  equals 900 – 1000 m/s, corresponding to  $p' = 3$  MPa.  $V_p$  increases  
249 slightly during the first minutes and then starts to increase quickly. The first period can be  
250 identified as the induction time that is necessary to create the first crystals of methane hydrates  
251 (Natarajan *et al.*, 1994). In this study, the induction time is approximately 0.2 h. After the  
252 induction time,  $V_p$  increases quickly during the next 10 h to progressively reach stabilization.  
253 The kinematic of  $V_p$  evolution within elapsed time is generally independent of the water  
254 saturation. The final value of  $V_p$ , however, depends on the initial water saturation and this

255 dependency will be analyzed later. It should be noted that the flow rate of the gas injection at  
256 the beginning was much higher than the limit of the flowmeter. For this reason, the total quantity  
257 of injected gas could not be measured by the flowmeter.

258

259 Figure 7 shows  $V_p$  versus elapsed time during the water saturation (Step 4). It should be noted  
260 that time zero corresponds to the beginning of the gas-water shift. For the reference test,  $V_p$   
261 increases when the sample is saturated with water. In contrast, for all tests with methane  
262 hydrates, replacing the excess methane gas with water decreases  $V_p$ .

263

264 As mentioned above, for the tests following procedure A, triaxial compression tests were  
265 performed after the water saturation (Step 4) at 22 MPa of confining pressure and 19 MPa of  
266 pore pressure, while for the tests following procedure B, a heating/cooling cycle was applied  
267 beforehand to dissociate and recreate the methane hydrates inside the sample. The results  
268 obtained during the heating path (procedure B) are shown in Figure 8. After decreasing the  
269 pore pressure from 7 MPa to 4 MPa, all the inlets were closed. Afterward, the sample was  
270 heated in undrained conditions inducing the increase in pore pressure. The pore pressure  
271 reached 19 MPa when the temperature of the cell reached 20°C. As mentioned above, when the  
272 pore pressure reached 19 MPa, the volume/pressure controller (8) was connected to the sample  
273 to maintain the pore pressure at 19 MPa, while the temperature of the cell continued to be  
274 increased. Figure 8c plots the volume of water (and/or methane gas) expelled during this phase.  
275 It varies from  $4 \times 10^{-6} \text{ m}^3$  to  $15 \times 10^{-6} \text{ m}^3$ , which corresponds to 5% and 20%, respectively, of the  
276 pore volume. In addition, the sample having the higher initial water saturation (higher hydrate  
277 saturation) shows a higher volume of expelled water.

278

279 Figure 9 presents pore pressure versus cell temperature during this heating path. The phase  
280 diagram (relationship between gas pressure and temperature during the dissociation of methane  
281 hydrates) is also plotted (Sloan & Koh, 2008). The results show that during this undrained  
282 heating path, the  $p - T$  plots closely follow the phase diagram (confirming the presence of  
283 methane hydrates in the system, after Kwon et al., 2008). However, at a given pressure, the cell  
284 temperature ( $T$ ) is slightly higher than the corresponding temperature determined by the phase  
285 diagram. This can be explained by the partial dissociation of hydrates and the continuous  
286 heating process (Figure 8a) where the cell temperature is slightly higher than the sample  
287 temperature (see also the schematic view of the cell in Figure 2). The  $p - T$  plots would be  
288 closer to the phase diagram if the heating rate was smaller.

289

290 After this dissociation step, to reform gas hydrates, the pore pressure was maintained at 19 MPa,  
291 while the cell temperature was decreased to 3 – 4°C (except for test B2). Figure 10 shows the  
292 cell temperature and the volume of water injected into the sample versus the elapsed time during  
293 this step. Except for the case of test B4, the volume of water injected into the sample is similar  
294 to that expelled (shown in Figure 8c) and reaches stabilization after 100 h. The volume  
295 measured in test B4 continues to increase with a constant rate even after 120 h. This was then  
296 attributed to a possible leakage in the connection that occurred due to the heating/cooling cycle.  
297 For test B2, the temperature was first decreased to 15°C, hydrate reformation started and  
298 became remarkable after 2 hours. The temperature was finally set at 3-4°C like in the other  
299 tests.

### 300 **3.2. Compressional wave velocity**

301 Figure 11 shows  $V_p$  at different steps during the methane hydrate formation process for all the  
302 tests. After the consolidation (Step 2),  $V_p$  equals to 900 – 1000 m/s for all the samples, as shown  
303 in Figure 5 (corresponding to  $p' = 3$  MPa). After the formation of methane hydrates under the

304 gas-saturated state (Step 3),  $V_p$  increases significantly (it equals 2500 – 3500 m/s). In addition,  
305 samples having similar water saturation show similar  $V_p$ . The subsequent water saturation (Step  
306 4) slightly decreases the  $V_p$  of the gas-saturated MHBS samples, while it increases the  $V_p$  of the  
307 reference sample. These values vary in the range of 1800 – 2600 m/s. After the gas hydrate  
308 dissociation phase, the  $V_p$  of all the four samples for B decreases and approaches the  
309 corresponding values obtained by the end of the consolidation (Step 2); they vary between 1000  
310 and 1200 m/s. Finally, the re-creation of gas hydrates increases  $V_p$  again to the range obtained  
311 before the dissociation step (end of Step 4), namely, between 1800 – 2300 m/s.

312

313 The results obtained at the end of the gas hydrate formation in the gas-saturated state (Step 3)  
314 are presented in Figure 12 where  $V_p$  is plotted versus hydrate saturation. The hydrate saturation  
315 was estimated from the initial water saturation and all the water was supposed to be used to  
316 create gas hydrates. It should be noted that this figure shows the results for eight tests, but the  
317 samples with the same moisture content have the same  $V_p$  (as shown in Figure 11). That reveals  
318 a good repeatability of the experimental procedure. These results show a clear effect of gas  
319 hydrate saturation on  $V_p$ .  $V_p$  is higher at a higher hydrate saturation; it increases from 900 –  
320 1000 m/s at the reference state (without hydrates) to 3500 m/s at  $S_h = 55\%$ .

321

322 In order to assess the gas hydrate distribution at the grain scale, the models proposed by  
323 Helgerud (2001) were used. To predict the gas-saturated MHBS morphology, Helgerud (2001)  
324 proposed two contact models using two different schemes: (i) gas hydrates are located only at  
325 the grain contacts (contact cement) and (ii) gas hydrates evenly envelop the grains (grain  
326 coating). The parameters used are shown in TTable 2. The results (Figure 12) show that the  
327 experimental data obtained in the present work correspond to the zone delimited by these two  
328 models. In addition, at low hydrate saturation ( $S_h = 27\%$ ) the experimental data fit the “grain

329 coating” model, while at high hydrate saturation ( $S_h = 55\%$ ), the “contact cement” model is  
330 more appropriate. The intermediate values of  $S_h$  correspond to the intermediate values of  $V_p$ .

331

332 The results corresponding to the water-saturated state are plotted in Figure 13. In this figure,  
333 the hydrate saturation was calculated from the quantity of methane gas dissociated at the end  
334 of each test (the gas that dissolved in water was ignored). The data obtained after the gas hydrate  
335 reformation, with the temperature cycle (only for procedure B), are plotted with closed symbols,  
336 while the data obtained without the temperature cycle (for procedure A tests) and before the  
337 temperature cycle (for procedure B tests) are plotted with open symbols. It should be noted that,  
338 for procedure B,  $V_p$  decreases after the temperature cycle in tests B2, B3 and B4, while it  
339 remains constant in test B1. Unlike the case of the gas-saturated state, the effect of hydrate  
340 saturation is less significant.  $V_p$  increases from 1850 m/s for the reference case (without  
341 hydrates) to 2500 m/s for  $S_h = 50\%$ . In addition, the  $V_p$  obtained after water saturation (Step 4)  
342 is generally higher than that obtained after the dissociation/recreation cycle at a given hydrate  
343 saturation.

344

345 In order to assess the gas hydrate distribution of water-saturated MHBS at the grain scale, four  
346 models proposed by Helgerud (2001) were used. In addition to the two contact models  
347 presented above, a model considering hydrates as fluid components (pore-filling) and another  
348 considering hydrates as sediment frame components (load-bearing) were used. The  
349 experimental and the models’ results are plotted in Figure 13 and show that after water  
350 saturation, the  $V_p$  data match the “load-bearing” model for some cases and are higher than the  
351 predicted value for other cases. After the temperature cycle (procedure B), the experimental  
352 data correspond to the pore-filling model, except for test B4 ( $S_h = 42\%$ ), where it is close to the  
353 load-bearing model.



### 3.3. Triaxial compressive properties

354  
355 The results obtained from the drained triaxial compression tests are shown in Figure 14 where  
356 deviator stress ( $q$ ) and volumetric strain ( $\varepsilon_v$ ) are plotted versus axial strain ( $\varepsilon_a$ ). It should be  
357 noted that all these samples were consolidated at 25 MPa of effective pressure prior to being  
358 unloaded to 3 MPa. The creation of methane hydrates and triaxial compression were performed  
359 at a mean effective stress of 3 MPa. The confining pressure and pore pressure were maintained  
360 at 22 MPa and 19 MPa, respectively, via the volume/pressure controller, while the axial strain  
361 rate was fixed at 0.1%/min. The volumetric strain was calculated based on the volume of water  
362 entering/being expelled from the sample. The results obtained from all the tests are quite  
363 similar: (i) the deviator increases almost linearly at the beginning, it reaches a peak value at  
364 about 2-3% of axial strain and decreases progressively to reach a residual state; (ii) the  
365 volumetric strain shows small contraction at the beginning and then decreases significantly  
366 (dilatation) prior to reaching the residual state at high axial strain ( $\varepsilon_a$  larger than 10%) where  $\varepsilon_v$   
367 remains constant. There is almost no difference between the results of the two procedures.

368  
369 The triaxial compressive properties, determined from the results shown in Figure 14, are plotted  
370 in Figure 15 versus hydrate saturation ( $S_h$ ). The peak strength,  $q_{max}$ , corresponds to the maximal  
371 value of the deviator (Figure 15a); the residual strength,  $q_{res}$ , corresponds to the deviator at the  
372 end of the test (Figure 15b). The secant Young's modulus,  $E_{50}$ , corresponds to the secant  
373 stiffness at 50% of the stress difference (Figure 15c). The dilation angle,  $\psi$ , is determined from  
374 the change in volumetric strain with respect to the change in shear strain by supposing that it is  
375 constant between 2 and 4% of the axial strain (Figure 15d). It is obvious that all these values  
376 are higher at a higher methane hydrate saturation.

377

378 Figure 16 shows an examination of all test results using Rowe's stress-dilatancy analysis for  
379 axial deformations before the peak strength was reached. For triaxial compression test  
380 conditions, the model is given by

$$381 \quad \frac{\sigma'_1}{\sigma'_3} = \tan^2 \left( \frac{\pi}{4} + \frac{\varphi_{cs}}{2} \right) \left( 1 - \frac{\dot{\varepsilon}_v}{\dot{\varepsilon}_1} \right)$$

382 where  $\sigma'_1$  and  $\sigma'_3$  are the major and minor effective stresses, respectively,  $\dot{\varepsilon}_v$  and  $\dot{\varepsilon}_1$  are the  
383 volumetric and major principal strain rates, respectively, and  $\varphi_{cs}$  is the critical state friction  
384 angle (Pinkert, 2017a).

385 All the experimental data are close to the values predicted by the model with  $\varphi_{cs} = 25^\circ$ , except  
386 for tests A3 and A4.

#### 387 **4. Discussion**

388 The induction time observed, approximately 0.2 h, is almost independent of the initial moisture  
389 content. This is in agreement with the results of Chong et al. (2016) who found an induction  
390 time of 0.1-0.8 h for fine sand (0.1 – 0.5 mm) prepared at 75% water saturation. Bagherzadeh  
391 et al. (2011) used the magnetic resonance imaging technique to investigate the formation of  
392 methane hydrates in an unconsolidated bed of silica sand and found that hydrate formation is  
393 not uniform and that the nucleation of hydrate crystals occurs at different times and different  
394 positions inside the bed. In addition, the hydrate formation was found to be faster with a lower  
395 moisture content and a smaller particle size. In the present work, only one particle size  
396 distribution was considered and the water saturation varied in a small range (from 25 to 50%).  
397 That would explain the independency of the induction time on the moisture content.

398

399 Figure 12 shows that gas hydrates significantly increase the compressional wave velocity at the  
400 gas-saturated state. The observation of the water distribution in the unsaturated sand at the pore

401 scale of Crist et al. (2004) reveals that water can be distributed around the grains as a thin layer  
402 (by adsorption) and also at the menisci of the grain contacts (by capillarity). In addition, at low  
403 water saturation, the water is mainly distributed at the surface of the grains (because the  
404 adsorption suction is much higher than the capillary suction); the volume of water at the menisci  
405 becomes dominant only at high water saturation. This was also observed by Riedel et al. (2012)  
406 via X-Ray Tomography at various water saturations. The injection of gas hydrates at 7 MPa of  
407 pressure and 3 – 4°C of temperature induces the transformation of water into the gas hydrates.  
408 The results show that this hydrate formation in gas-saturated media increases the  $V_p$  (see Figure  
409 6), and this increase reaches the stabilization state after about 100 h. This stabilization suggests  
410 that almost the entire quantity of available water has been transformed into gas hydrates. In  
411 addition, the distribution of gas hydrates should be similar to that of water prior to its creation.  
412 As a consequence, at a low moisture content (hydrate saturation), the gas hydrates are mainly  
413 distributed around the grains, while at a high moisture content (hydrate saturation), the role of  
414 the gas hydrates at the grain contacts dominates. This explanation is in agreement with the  
415 comparison between the experimental data and Helgerud's model shown in Figure 12.  
416 Chaouachi et al. (2015) investigated the formation of Xenon gas hydrates in under-saturated  
417 sediments with juvenile water using synchrotron X-ray computed tomographic microscopy.  
418 The results showed that the nucleation started at the water-gas interface resulting in an initial  
419 gas hydrate film, several micrometers in thickness, and future growth proceeded to form  
420 isometric single crystals, 10 – 20 micron in size. In the study of Yoneda et al. (2016), the  
421 formation of krypton hydrates in partially saturated sand was observed using micro-focus X-  
422 ray computed tomography. The results also confirmed the formation of hydrates from the  
423 capillary bridges of wet sand and the patchy initial distribution of water.

424

425 After the formation of gas hydrates in gas-saturated state, the subsequent water saturation phase  
426 significantly decreases the  $V_p$  (Figure 7). Ebinuma et al. (2008) and Kneafsey et al. (2010)  
427 observed a similar decrease in sonic velocities when saturating the gas-saturated MHBS with  
428 water. Hyodo et al. (2013) compared the mechanical properties of gas-saturated MHBS with  
429 those of water-saturated ones (using the same sand under the same stress conditions) and found  
430 higher stiffness and higher failure strength for gas-saturated specimens. It should be noted that  
431 the procedure applied by Hyodo et al. (2013) to obtain water-saturated MHBS is similar to  
432 procedure A used in the present work. These results suggest that water saturation modifies the  
433 hydrate distribution at the grain scale; the gas hydrates located at the grain contacts would be  
434 progressively converted or/and redistributed into the pore spaces (Choi et al., 2014). In this  
435 study, the effect of water injection in gas-saturated hydrate-bearing sediments is investigated at  
436 different levels of hydrate saturation. The results shown in Figure 7 and Figure 13 indicate that  
437 this process takes longer for higher hydrate saturation. For some specimens (B2, B4, A3, and  
438 A4), this transformation was not complete when the subsequent step (triaxial compression for  
439 specimens A and heating/cooling cycle for specimens B) was applied. This can be explained  
440 by the higher values for  $V_p$  than the predicted values of the load-bearing model. In addition, for  
441 the two tests following procedure A (A3 and A4), even after waiting a long time for water  
442 saturation (to make sure that the MHBS was well saturated with water), the  $V_p$  were still higher  
443 than the values predicted with the load-bearing model. This can perhaps be explained by the  
444 patchy hydrate distribution suggested by Dai et al. (2012) where the bulk stiffness approaches  
445 the lower Voigt-Reuss bound at low hydrate saturation and the upper bound at high hydrate  
446 saturation due to the mechanical interaction between the hydrate-saturated patches. As  
447 aforementioned, the number of tests is not so high, but the procedure was correctly controlled  
448 in the laboratory. The repeatability of the tests can be appreciated from the results of the

449 consolidation step (Step 2), seen in Figure 4 and Figure 5, and the hydrate formation in gas-  
450 saturated media (Step 3), seen in Figure 6.

451

452 In the present work, methane gas was injected into an unsaturated sand specimen to create a  
453 MHBS sample. Afterward, water was injected to saturate the specimen. This procedure is  
454 similar to that used by Miyazaki et al. (2011) and Hyodo et al. (2013) to investigate the triaxial  
455 compressive properties of artificial MHBS. As the initial distribution of water inside the  
456 specimen should be homogenous (partly due the equilibrium of the capillary force), it was  
457 expected that the distribution of gas hydrates inside the sample would also be homogenous  
458 within this method. However, Kneafsey et al. (2010), using X-ray computed tomography,  
459 found that this method produced both well-distributed hydrates and very heterogeneous  
460 hydrates. They noted that the heterogeneity in the hydrate saturation (observed in the work of  
461 Seol & Kneafsey, 2009) was affected by the initial sample heterogeneity, but not dominated by  
462 it. In the present work, as the sand specimen was well compacted, the specimen density was  
463 relatively homogenous. As a result, the gas hydrate distribution is expected to be homogenous.  
464 Observation by magnetic resonance imaging (MRI) on a sample prepared by the same  
465 procedure has confirmed this statement (see Le et al., 2019).

466

467 The subsequent heating/cooling cycle aims at dissociating/recreating gas hydrates without  
468 disturbing the homogeneity of the hydrate distribution. For this reason, during the heating path,  
469 the drainage system was closed during the first period. This condition induced an increase in  
470 pore pressure (Figure 8 and Figure 9) mainly due to thermal dilation of water and hydrate  
471 dissociation (Kwon et al., 2008). The undrained heating condition was stopped when the pore  
472 pressure approached the limit of the pore pressure transducer (20 MPa). A small quantity of  
473 water (and dissolved methane gas) was then allowed to be expelled. As the final state (in terms

474 of pore pressure and temperature) is far outside the phase diagram (Figure 9), the methane  
475 hydrates should be completely dissociated at the end of the heating path. During the subsequent  
476 cooling path, the pore pressure was maintained at 19 MPa, and the cooling induced the injection  
477 of water to the specimen (Figure 10). This corresponds mainly to both the thermal contraction  
478 of water and the gas hydrate reformation. The reformation of gas hydrates can be easily  
479 identified when the cell temperature remains constant, while the volume of injected water  
480 increases abruptly (e.g., at 12 h for test B2).

481

482 Choi et al. (2014) formed methane hydrates with the partial water saturation method (excess  
483 gas method), injecting saline water at locations just outside of the hydrate stability zone for  
484 saline water and doing a temperature cycle. It was concluded that slow saline water injection is  
485 the key to initiating the formation of non-cementing hydrates and that the temperature cycle  
486 ensured this formation. The  $V_p$  after warming was quite high, while the sample was not  
487 saturated. The hydrate dissociation was perhaps not completed before the hydrate reformation.  
488 This explains why the difference in  $V_p$  between after the saline water injection and after the  
489 temperature cycle was not obvious. The measurement of  $V_p$  at the end of the thermal cycle in  
490 the present study is smaller than that obtained after water saturation. In addition, it fits with the  
491 pore-filling and load-bearing models. It can then be expected that the heating/cooling cycle  
492 allows for the completion of the conversion of the hydrate accumulation habit into the non-  
493 cementing type.

494

495 The experimental results obtained from the triaxial compression tests show higher values for  
496 the maximum deviator stress, secant Young's modulus, residual deviator stress, and dilation  
497 angle at a higher hydrate saturation. Hyodo et al. (2013) and Miyazaki et al. (2011) found a  
498 similar effect for the hydrate saturation on the maximum deviator stress and secant Young's

499 modulus while testing the MHBS prepared with a procedure similar to procedure A in the  
500 present work. The effect of the methane hydrate saturation on the stiffness and failure strength  
501 of MHBS was explained by particle bonding. However, the measurement of  $V_p$  in the present  
502 work suggests that grain-contact hydrates have been significantly converted into non-grain-  
503 contact (non-cementing) types by the end of procedure A, as explained above. In addition, the  
504 results obtained by procedure B (where the conversion has already been completed) show a  
505 similar effect of the hydrate saturation on the stiffness and failure strength of MHBS. Soga et  
506 al. (2006) showed a similar increase in the dilation angle with an increase in the hydrate  
507 saturation for natural samples and synthetic samples (partial water saturation method for strong  
508 grain contacts and ice-seeding method for weak grain contacts) at 1 MPa of confining pressure.  
509 It is obvious that an increase in the hydrate saturation enhances the dilative characteristics of  
510 soils, and this increase is more apparent when the hydrate saturation exceeds 30%. Recently,  
511 Pinkert (2017a,b) analyzed the experimental data obtained by a procedure similar to procedure  
512 A and also concluded that no cohesive effect was found when interpreting the test results under  
513 a stress-dilatancy framework.

514

515 In the present work, the cohesion and friction angle cannot be determined because only one  
516 level of effective confining pressure was used. For this reason, the approach of Pinkert  
517 (2017a,b) can be appropriated to give more information about the cohesion. When using the  
518 method proposed by Pinkert (2017a) to analyze the data obtained in the present work (Figure  
519 16), the results from tests A3 and A4 are positioned above the other curves. These results then  
520 correspond to a significant increase in the degree of cohesion in the samples. This is also in  
521 agreement with the results shown in Figure 13 and the above comments in which it is expected  
522 that the conversion of gas hydrates from grain contacts to pore spaces is not completed for these  
523 two cases. To simulate natural hydrate-bearing sediments, with a range in hydrate saturation of

524 0-50%, procedure A (without the temperature cycle) could be used when the hydrate saturation  
525 is smaller than 40%, while at higher saturation (40-50%), a temperature cycle should be added  
526 (procedure B should be used).

## 527 **Conclusions**

528 Methane hydrate-bearing sand was first created by pressurizing methane gas (at 7 MPa) into an  
529 already chilled moistened packed sandy specimen (excess gas method). Following the hydrate  
530 formation, water was injected into the sample and the remaining gas was simultaneously bled  
531 out. The water pressure was then maintained at 19 MPa (22 MPa of confining pressure) until  
532 water was no longer injected into the sample. That corresponds to the end of procedure A for  
533 preparing the MHBS specimen. For procedure B, a subsequent heating/cooling cycle was  
534 applied in order to completely dissociate the gas hydrates and then recreate them inside the  
535 specimen. Measurement of the compressional wave velocity was performed along with these  
536 processes, while triaxial compression tests were performed at the end of each procedure. Rock  
537 physics models were also compared to the experimental data to assess the grain scale  
538 distribution of the gas hydrates at each state. The following conclusions can be drawn:

- 539 - Pressurizing methane gas into an already chilled moistened packed sandy specimen  
540 creates gas hydrates at the grain contacts. This excess gas method allows for a  
541 significant increase in  $V_p$ .
- 542 - Subsequent water saturation converts (and/or redistributes) the gas hydrates from grain  
543 contacts to pore spaces and, as a result, decreases  $V_p$ . This process may take several  
544 days, depending on the hydrate saturation, and cannot be completed for high hydrate  
545 saturation.
- 546 - The heating/cooling cycle allows for the completion of the conversion (and/or  
547 redistribution) of gas hydrates from grain contacts to pore spaces.



548 - The effects of hydrate saturation on the mechanical properties of MHBS obtained by  
549 the two procedures (with and without thermal cycles) are similar at low hydrate  
550 saturation.

551 - The effect of the grain scale gas hydrate distribution can be detected in the case where  
552 the conversion (and/or redistribution) of gas hydrates from grain contacts to pore spaces  
553 is not complete.

554 As the non-cementing habit is actually the most representative case of natural gas hydrate-  
555 bearing sandy sediments, the findings of the present work would be helpful for future studies  
556 on gas hydrate-bearing sediments in the laboratory. Actually, within procedure A, non-  
557 cementing hydrates could be obtained by waiting long enough for the completion of the hydrate  
558 conversion from grain contacts to pore spaces in the case of small hydrate saturation. With the  
559 heating/cooling cycle (procedure B), non-cementing hydrates could even be obtained at higher  
560 hydrate saturation.

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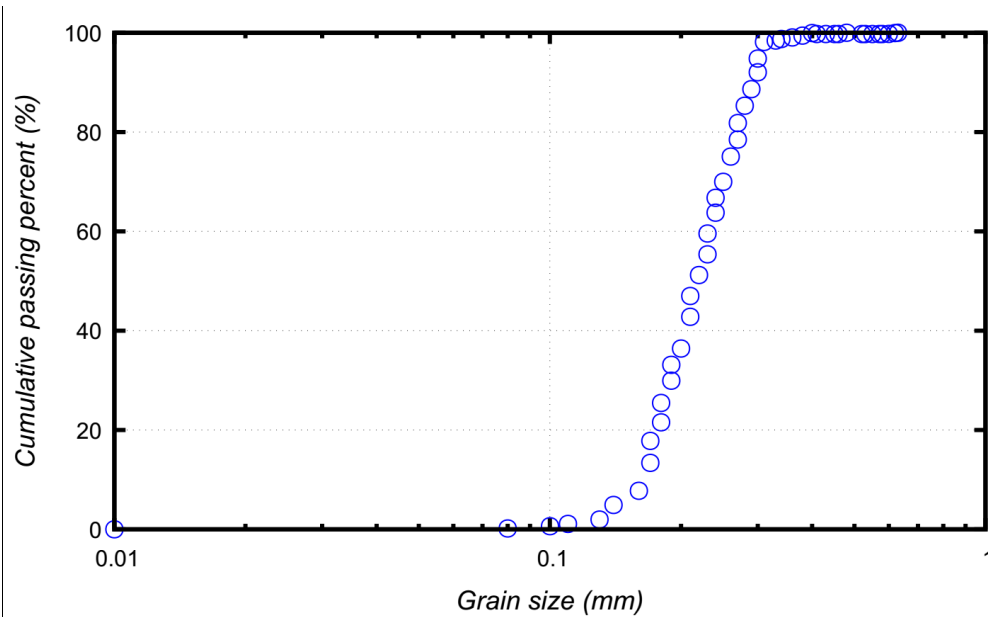


**Table 1. Test program**

<b>Test</b>	<b>Moisture content (%)</b>	<b>Water saturation (%)</b>	<b>Hydrate saturation (%)</b>
<i>Reference</i>	0.0	0	0
<i>A1</i>	6.0	25	21
<i>B1</i>	6.0	25	13
<i>A2</i>	8.5	35	31
<i>B2</i>	8.5	35	34
<i>A3</i>	10.0	42	50
<i>B3</i>	10.0	42	41
<i>A4</i>	12.0	50	48
<i>B4</i>	12.0	50	42

**Table 2. Parameters used for Helgerud's model**

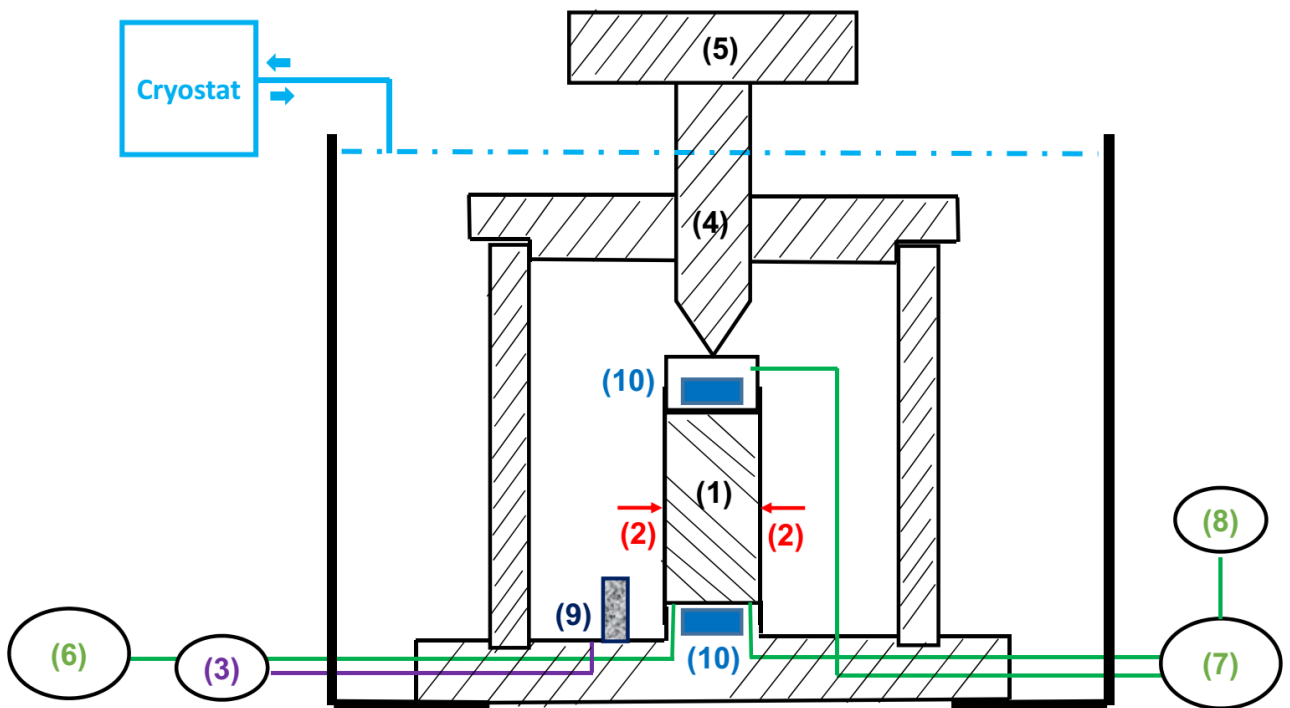
Parameter	Value
Bulk modulus of quartz (GPa)	36.6
Shear modulus of quartz (GPa)	45
Bulk modulus of water (GPa)	2.15
Bulk modulus of air (GPa)	0.01
Porosity (-)	0.387
Density of solid grain (Mg/m <sup>3</sup> )	2.65
Density of water (Mg/m <sup>3</sup> )	1
Density of air (Mg/m <sup>3</sup> )	0
Density of methane hydrate (Mg/m <sup>3</sup> )	0.9
Bulk modulus of hydrate (GPa)	7.9
Shear modulus of hydrate (GPa)	3.3
Critical porosity (-)	0.387
Number of contacts per grain (-)	4.5



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**Figure 1. Particle size distribution curve (modified from Feia et al., 2015)**

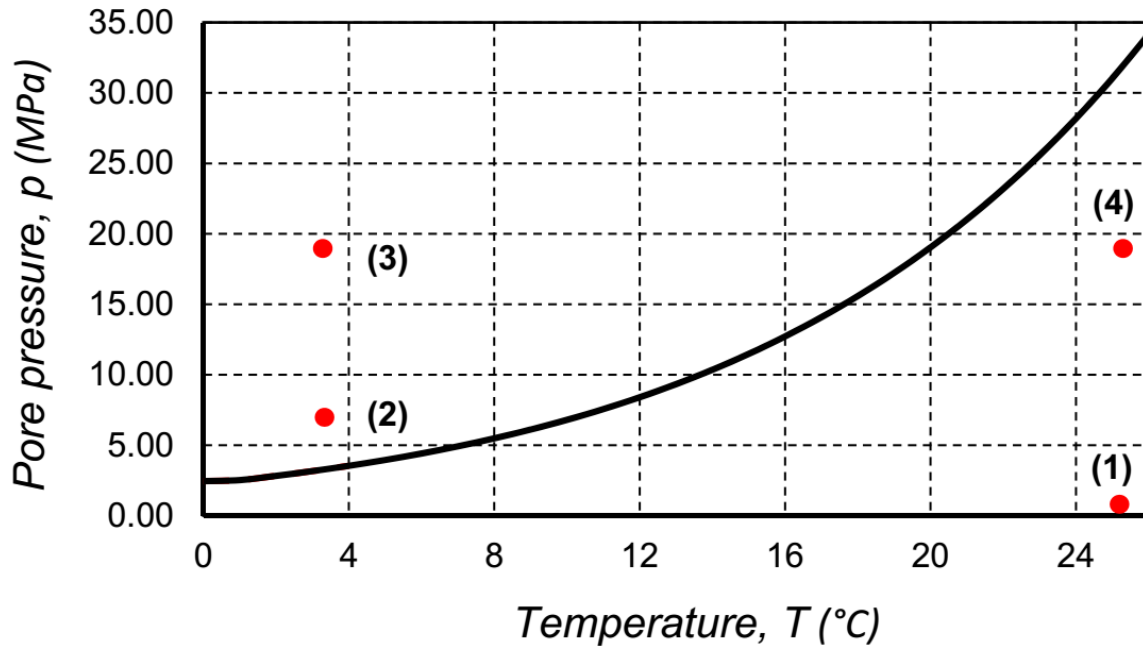


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**Figure 2. Schematic diagram of the experimental setup**

729 (1): sample; (2): displacement sensor; (3): volume/pressure controller to control confining  
 730 pressure; (4): piston; (5): force transducer; (6): pressure transducer connected to gas flowmeter  
 731 for methane gas injection; (7): top and bottom pore pressure transducers connected by a T valve;  
 732 (8): volume/pressure controller to control pore pressure (water pressure); (9): thermocouple;  
 733 and (10): two ultrasonic sensors connected to a wave generator for measurement of  $V_P$ .



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**Figure 3.  $p$ - $T$  conditions during gas hydrate formation**

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(1): Initial state of the sample (Step 1 and Step 2);

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(2):  $p$ - $T$  during GH formation in gas-saturated media (Step 3) and after water saturation (Step 4);

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(3):

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- Procedure A:  $p$ - $T$  at the end of water saturation when confining pressure and pore pressure were increased to 22 MPa and 19 MPa, respectively;

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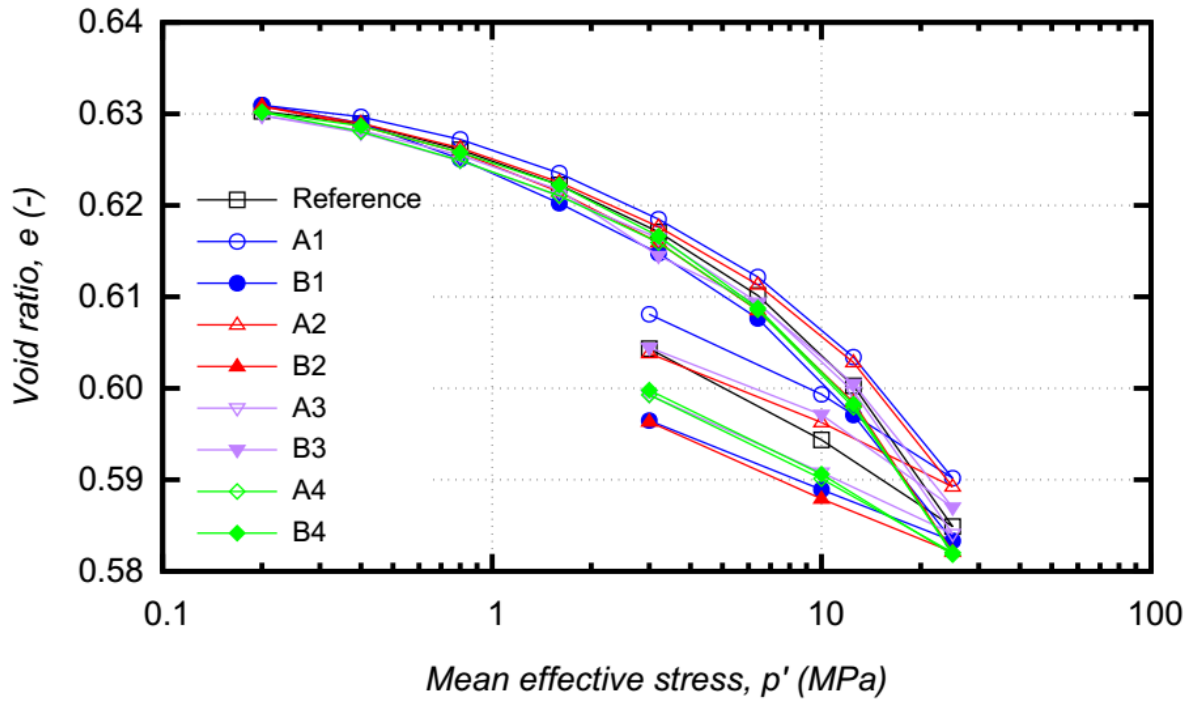
- Procedure B:  $p$ - $T$  during gas hydrate reformation;

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(4):  $p$ - $T$  at the end of gas hydrate dissociation for procedure B tests.

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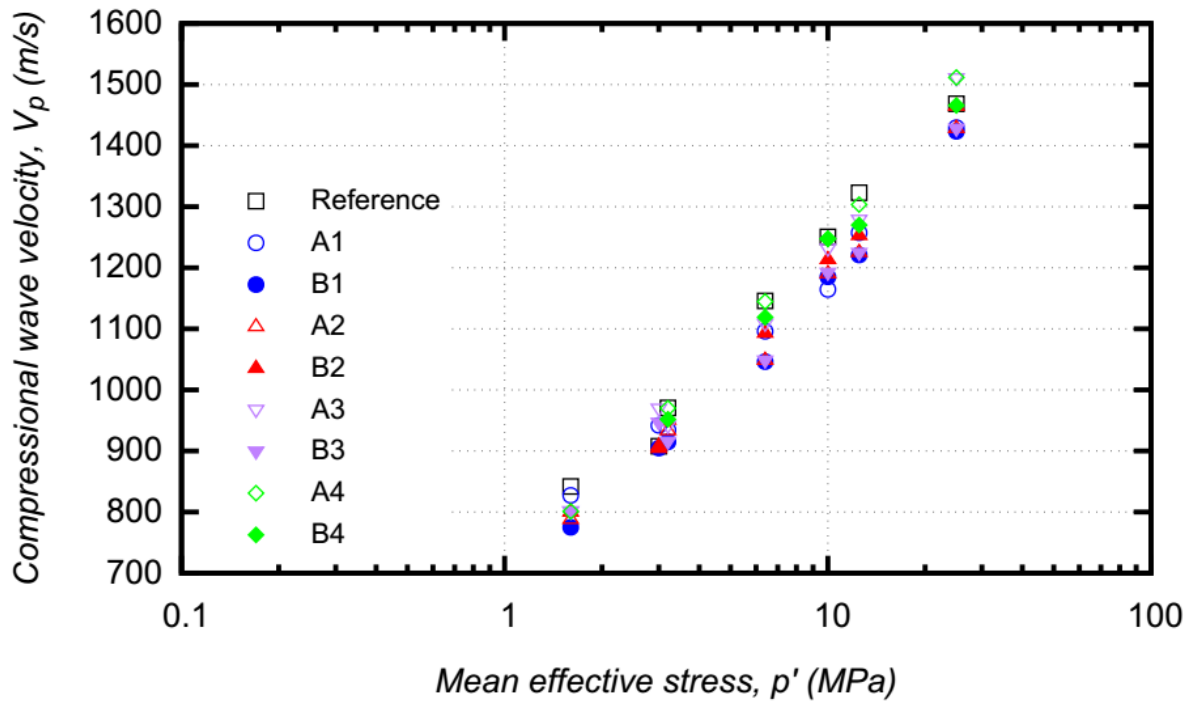
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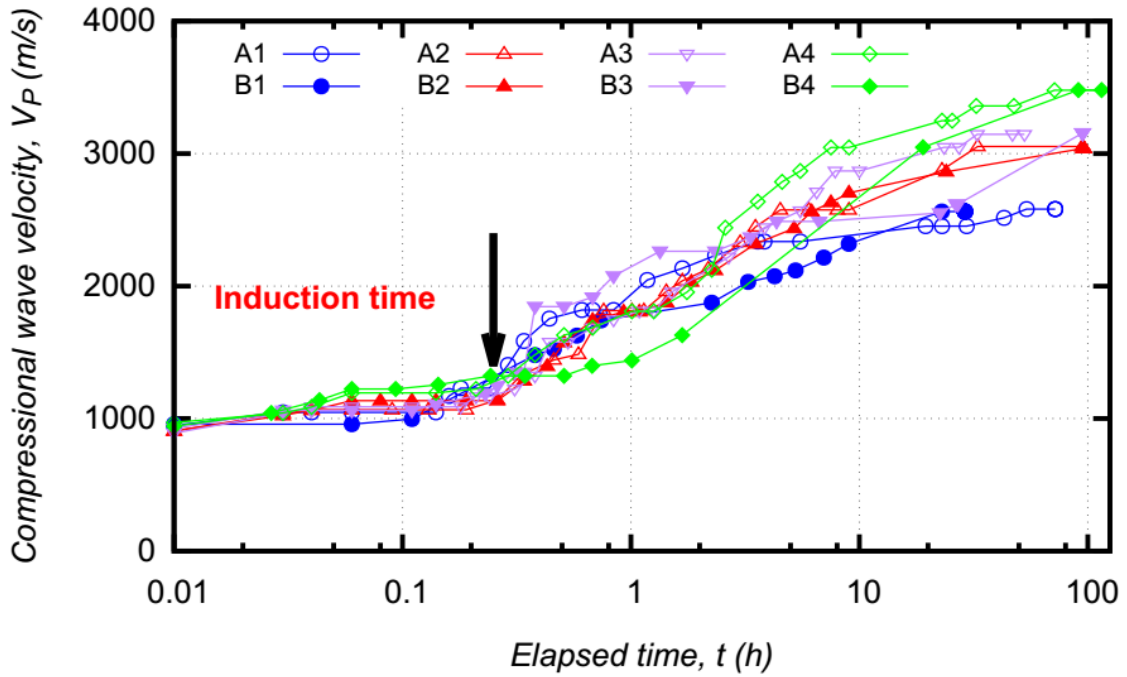
Figure 4. Void ratio versus effective stress for all tests



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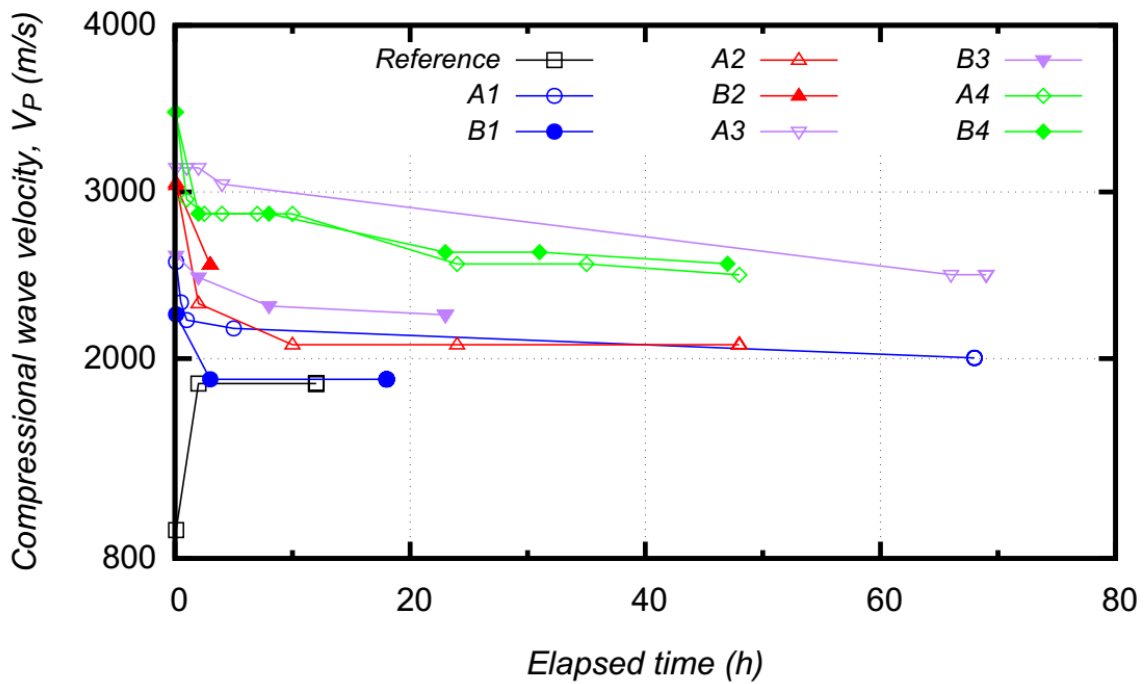
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Figure 5. Compressional wave velocity versus effective stress for all tests



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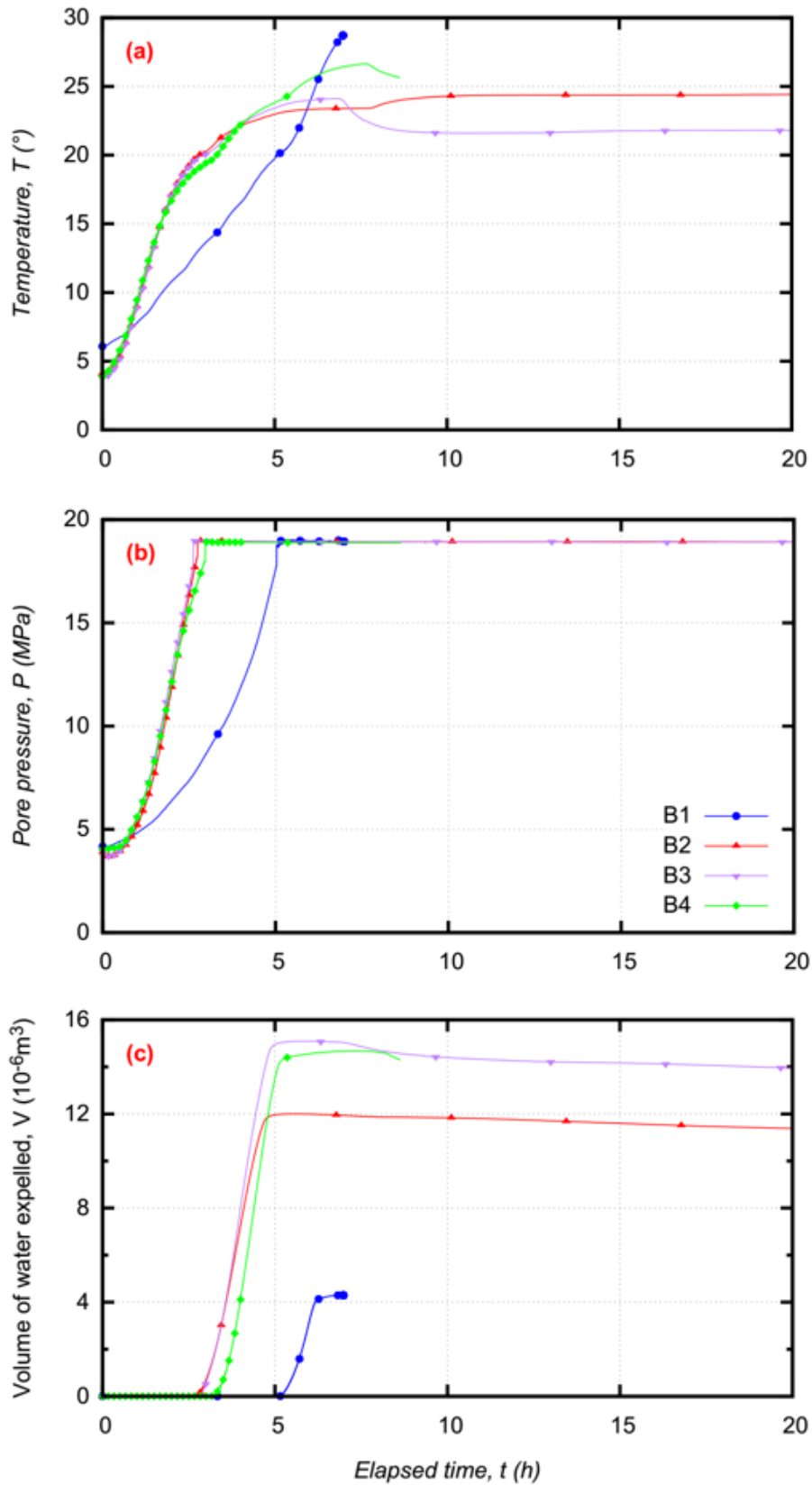
751 **Figure 6. Compressional wave velocity versus elapsed time during methane hydrate**  
 752 **formation in gas-saturated media**



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754 **Figure 7. Compressional wave velocity versus elapsed time during water saturation**

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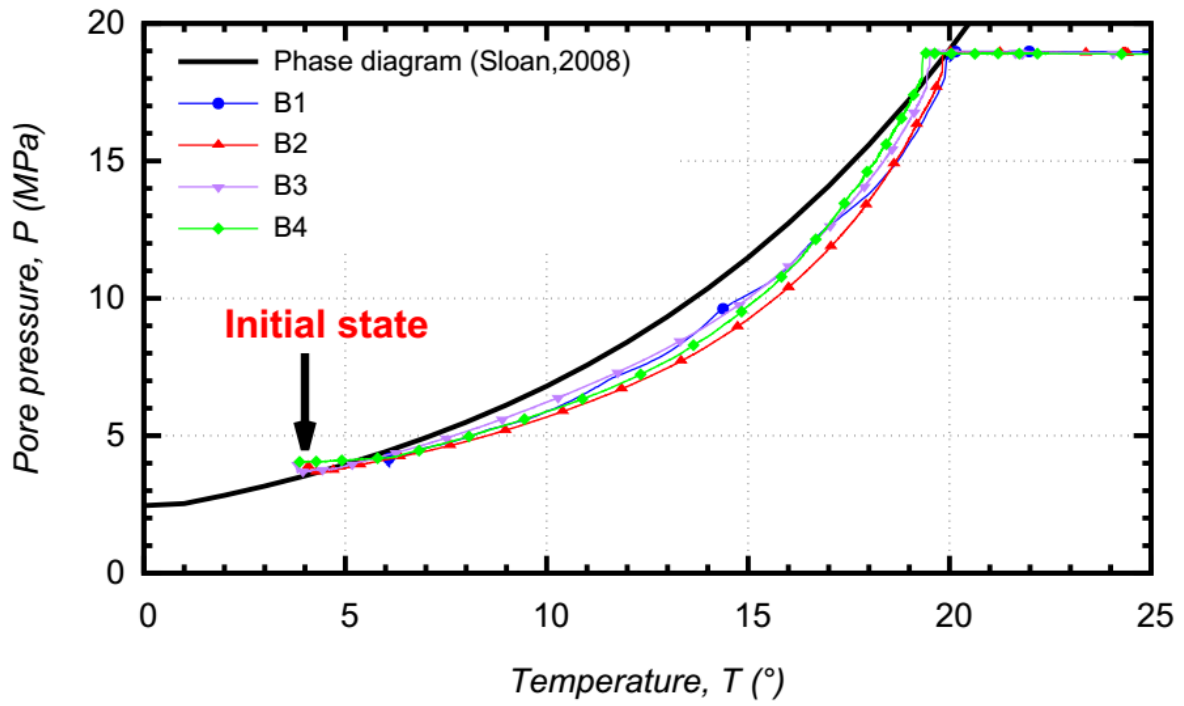


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757 **Figure 8. Pore pressure, temperature and volume of water expelled versus elapsed time**  
 758 **during heating path**

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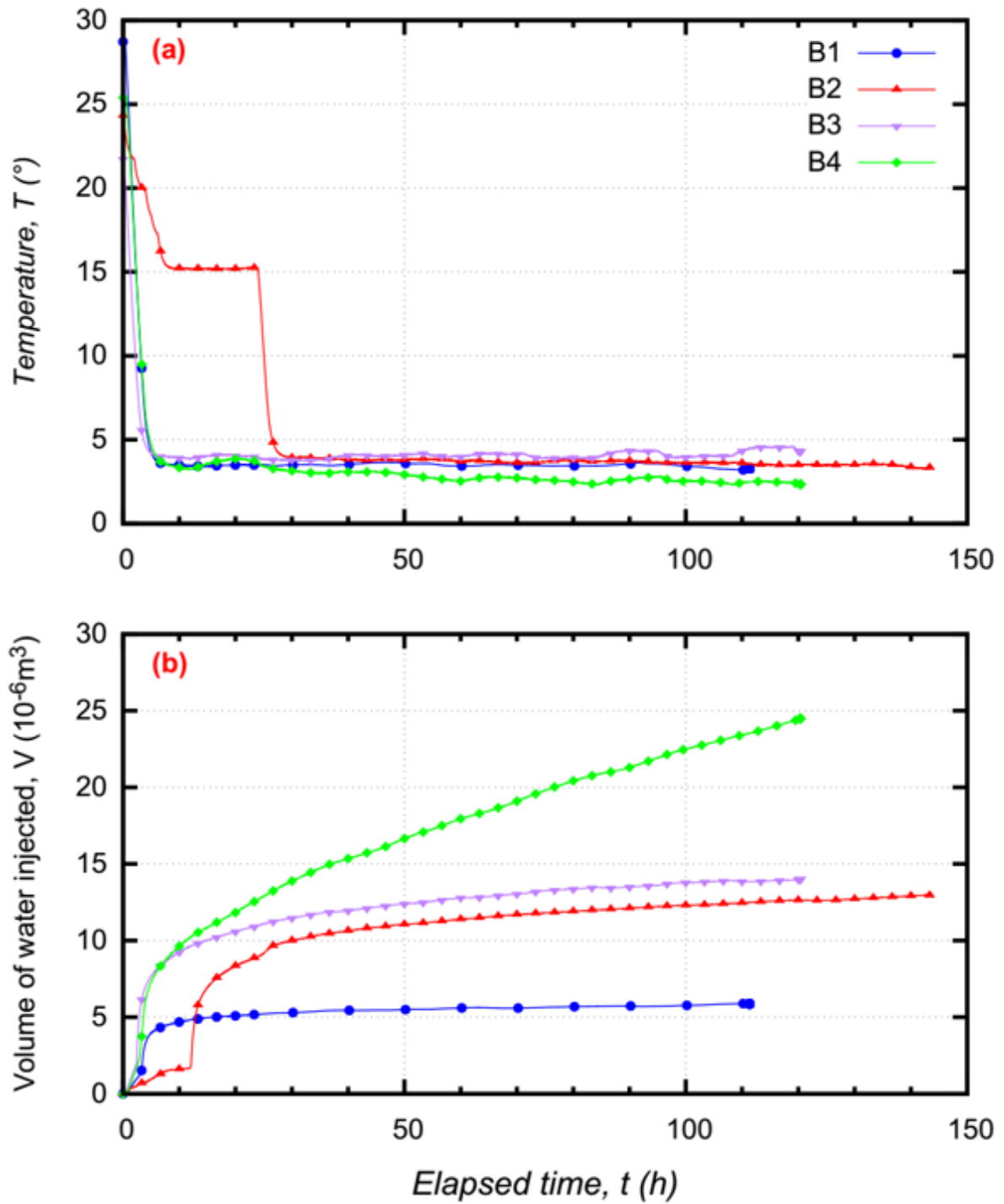
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**Figure 9. Pore pressure versus cell temperature during dissociation**

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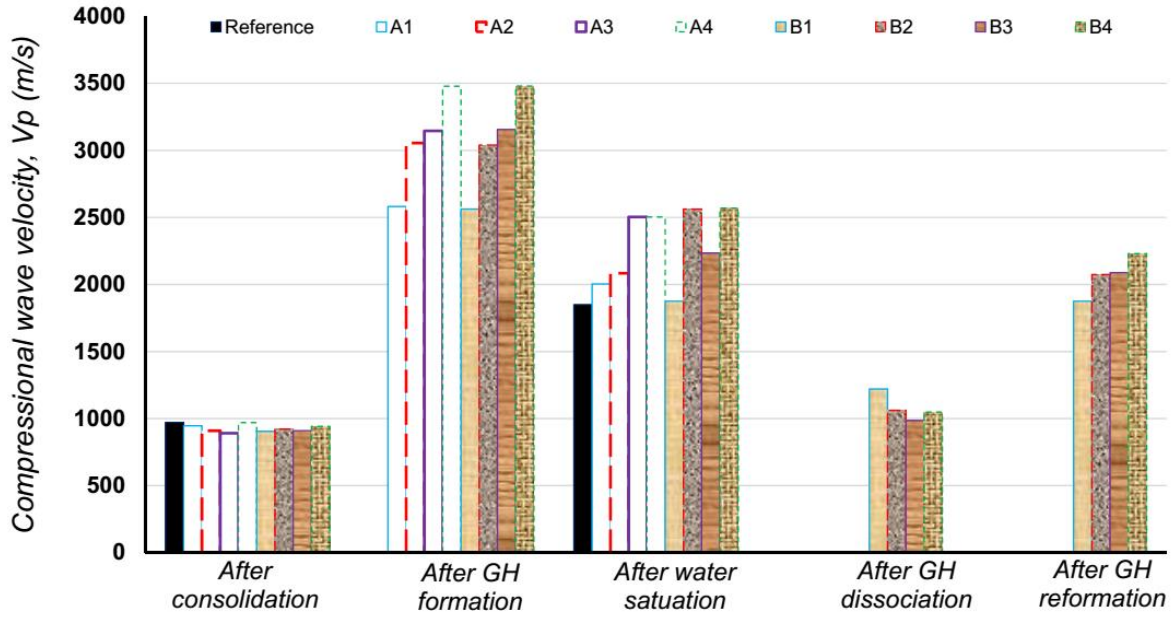
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 766 **Figure 10. Temperature and volume of injected water versus elapsed time during**  
 767 **hydrate reformation**

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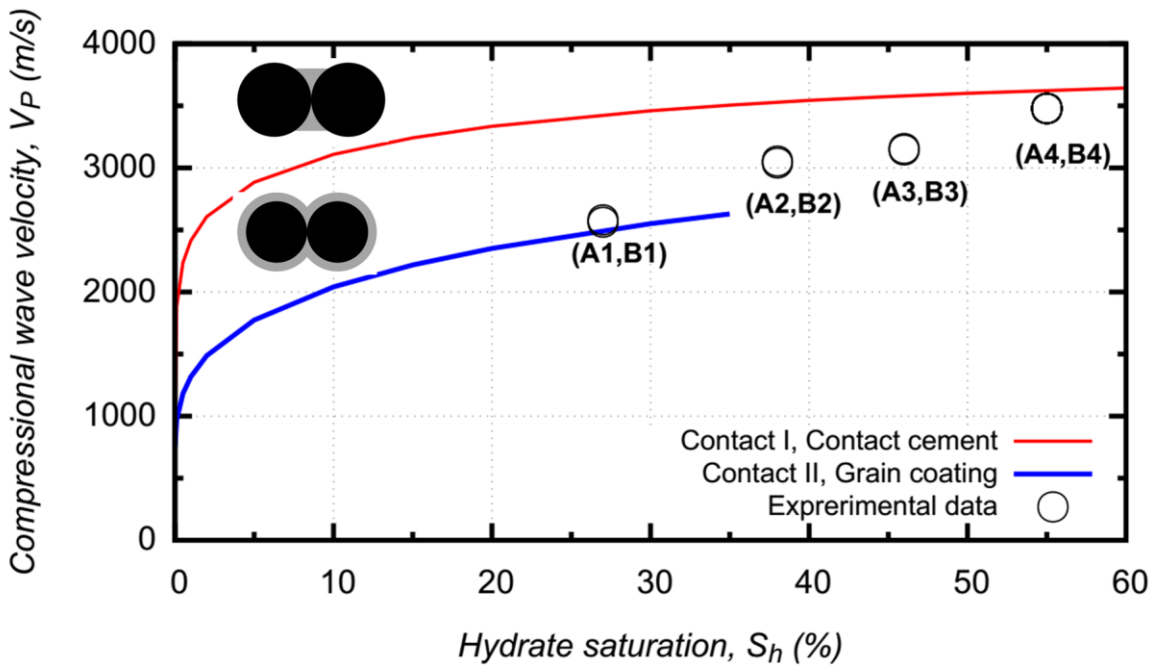




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770 **Figure 11. Compressional wave velocity during the whole methane hydrate formation of**  
 771 **all tests**

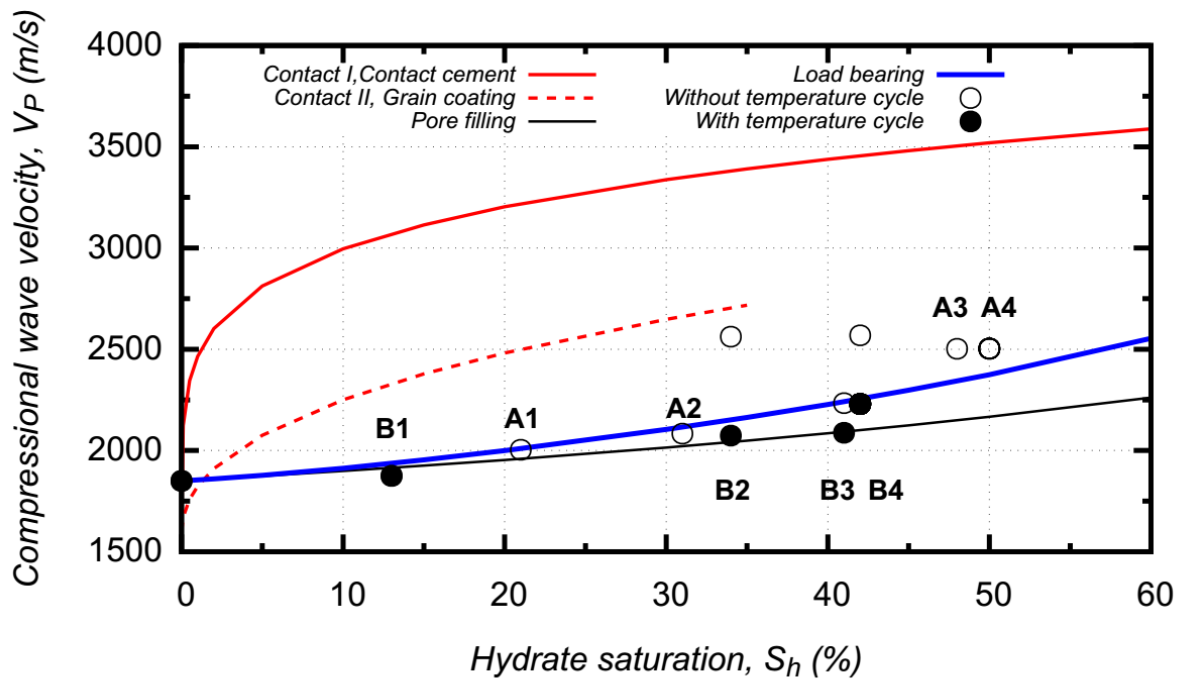
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774 **Figure 12. Comparison between experiments and Helgerud's model of compressional**  
 775 **wave velocity dependence on methane hydrate saturation in gas-saturated media**

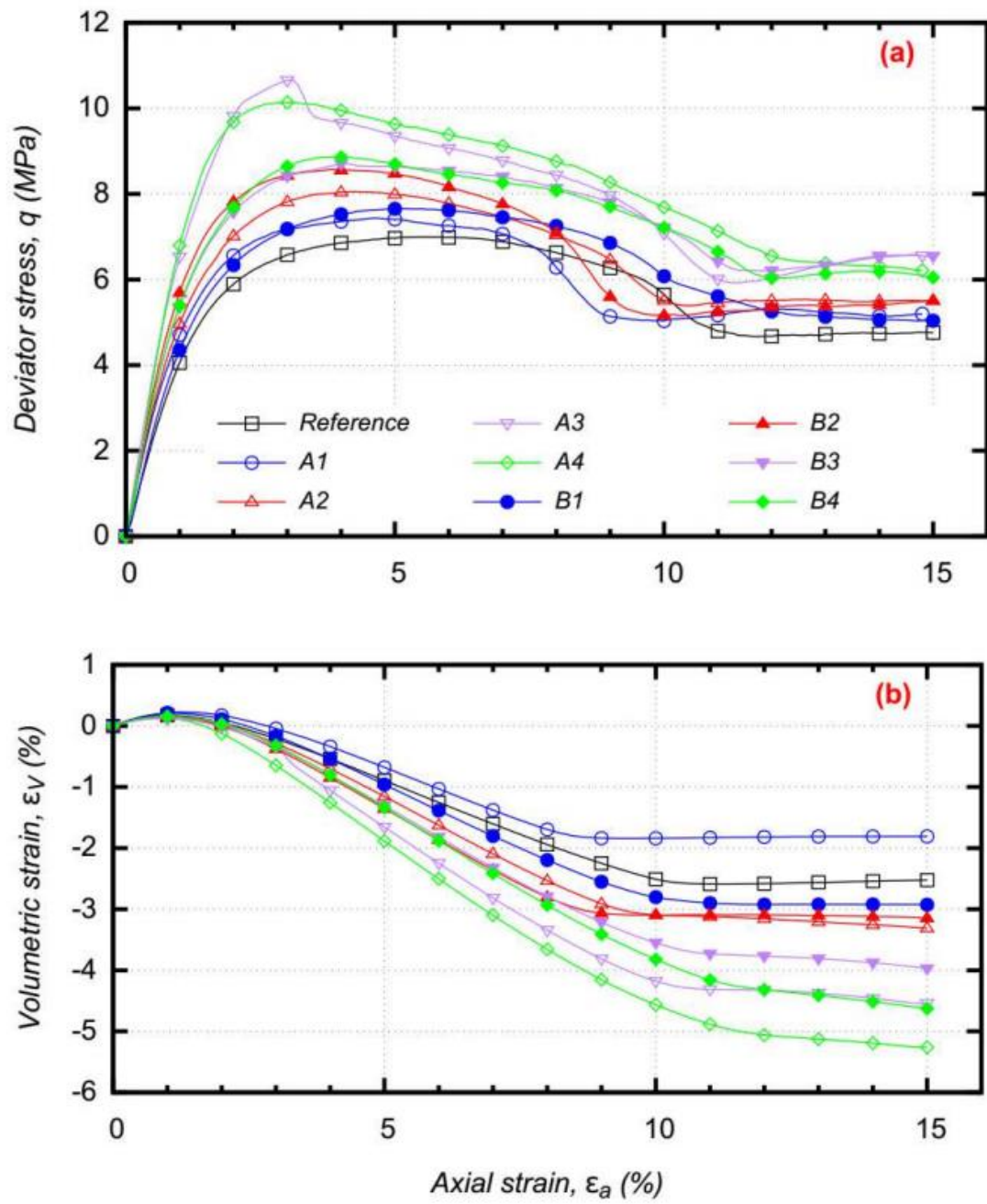
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**Figure 13. Comparison between experiments and Helgerud's model of compressional wave velocity dependence on methane hydrate saturation in water-saturated media**

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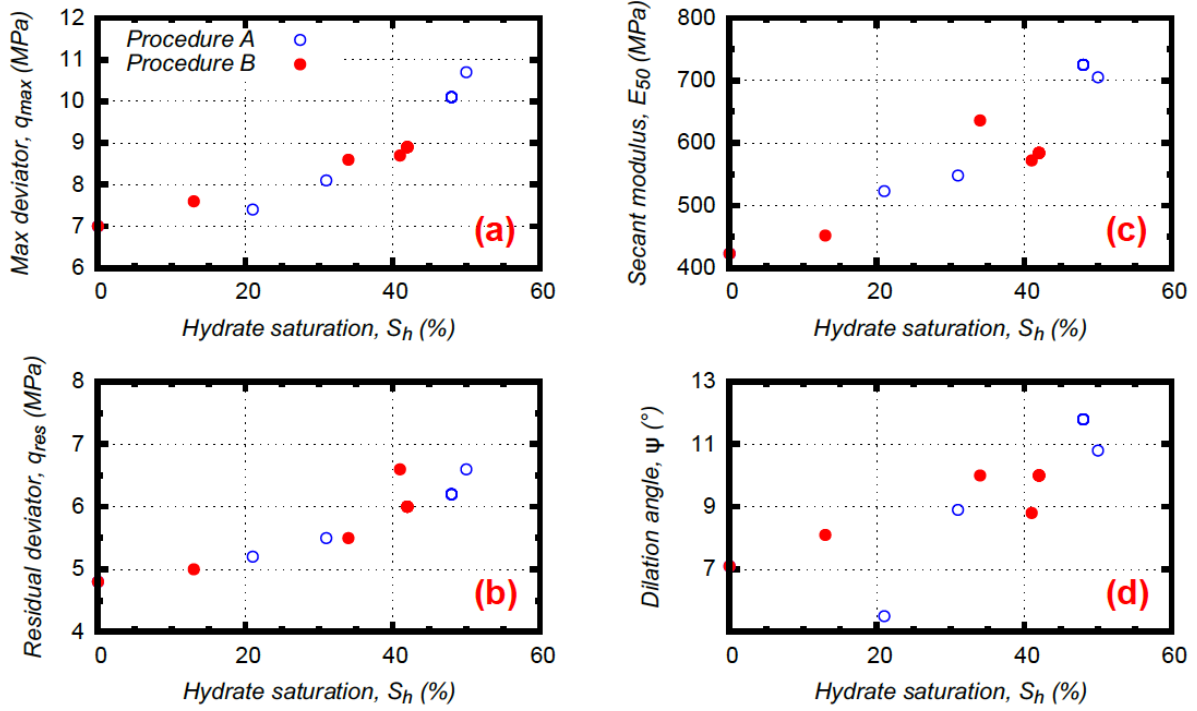


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Figure 14. Deviator and volumetric strain versus axial strain

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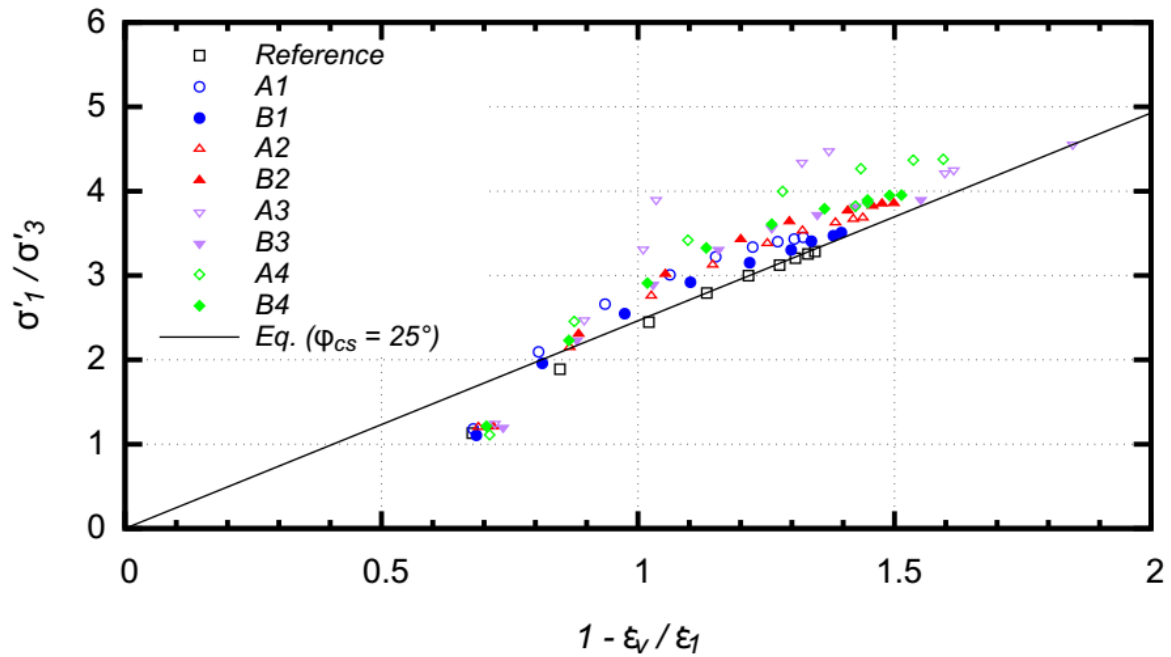


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**Figure 15. Dependence of mechanical properties of sand on methane hydrate saturation for all tests**



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**Figure 16.  $\sigma'_1/\sigma'_3$  versus  $1 - \epsilon_v/\epsilon_1$**

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