

# A densification mechanism to model the mechanical effect of methane hydrates in sandy sediments

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## SUMMARY

Recent pore-scale observations and geomechanical investigations suggest the lack of true cohesion in methane hydrate-bearing sediments (MHBS) and propose that their mechanical behavior is governed by kinematic constrictions at pore-scale. This paper presents a constitutive model for MHBS, which does not rely on physical bonding between hydrate crystals and sediment grains, but on the densification effect that pore invasion with hydrate has on the sediment mechanical properties. The Hydrate-CASM extends the critical state model Clay and Sand Model (CASM) by implementing the subloading surface model and introducing the densification mechanism. The model suggests that the decrease of the sediment available void volume during hydrate formation stiffens its structure and has a similar mechanical effect as the increase of sediment density. In particular, the model attributes stress-strain changes observed in MHBS to the variations in sediment available void volume with hydrate saturation and its consequent effect on isotropic yield stress and swelling line slope. The model performance is examined against published experimental data from drained triaxial tests performed at different confining stress and with distinct hydrate saturation and morphology. Overall, the simulations capture the influence of hydrate saturation in both the magnitude and trend of the stiffness, shear strength and volumetric response of synthetic MHBS. The results are validated against those obtained from previous mechanical models for MHBS that examine the same experimental data. The Hydrate-CASM performs similarly to previous models but its formulation only requires one hydrate-related empirical parameter to express changes in the sediment elastic stiffness with hydrate saturation.

**KEY WORDS:** methane hydrate-bearing sediments; mechanical behavior; densification mechanism; Hydrate-CASM; constitutive modeling.

## 1. INTRODUCTION

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Methane hydrates have drawn international interest as an alternative energy resource to conventional fossil fuels [1-5], and as a major hazard for offshore drilling and gas production operations [6-8], global climate change [9-12] and seafloor instability [13-15]. Quantitative evaluation of the resource potential of gas hydrate reservoirs and of their response to natural and/or human-induced changes in pressure and temperature (P-T) conditions, requires precise knowledge of the hydrate phase change phenomenon and of its effect on the mechanical stability of the reservoir. Due to the operational complexity at preserving the in-situ P-T conditions during MHBS recovery, the mechanical properties of these sediments are generally investigated through geophysical techniques [16-18] and geotechnical testing of synthetic sediments [19-21]. Both geophysical and geotechnical data show that the stiffness, strength, and dilatancy of MHBS tend to increase with increasing hydrate saturation [22, 23]. They also evidence that their mechanical and hydraulic properties drastically change during hydrate dissociation, which may compromise the mechanical stability of the sediment. Thus, hydrate dissociation is likely to trigger small to large-scale deformations in the seabed, including sediment collapse [24] and sliding [25-27]. As a result, dissociation may also induce damage of preexisting offshore infrastructures [28].

Several mechanical models developed for MHBS assume that the increase of strength, stiffness and dilatancy observed in these sediments is mainly governed by bonding or cementation between the hydrate crystal and the sediment grains (Table 1). However, recent pore-scale observations [29-31] and geomechanical investigations [32-34] evidence the lack of true cohesion in MHBS and suggest that the mechanical response of these sediments may not necessarily be governed by sediment bonding/cementation, but rather to kinematic constrictions at pore/grain scale during shearing. In this paper, we develop a new mechanical constitutive model that does not consider hydrate-bonding effects in its formulation but assumes that the reduction of sediment available void volume and the increase of sediment elastic stiffness during pore invasion with hydrate can explain the greater mechanical properties observed in MHBS

The elasto-plastic model Hydrate-CASM extends the formulation of the unified critical state constitutive model CASM [46] by implementing the subloading surface model [47] and introducing the densification mechanism. The subloading surface, which has been successfully

69 used in previous mechanical models for MHBS [39, 40, 44, 48], allows capturing irrecoverable  
70 plastic strains inside the yield surface. The densification mechanism suggests that the decrease  
71 of the available void volume of the host sediment during hydrate formation stiffens its structure  
72 and has a similar mechanical effect as the increase of the sediment density. In particular, the  
73 densification mechanism attributes the stress-strain changes observed in MHBS to variations  
74 in sediment available void volume with hydrate saturation and its consequent effect on isotropic  
75 yield stress and swelling line slope.

76

77 The Hydrate-CASM is applied here to robust and well-described published experimental data  
78 [19, 21] that cover the most relevant conditions related to MHBS behavior, including a wide  
79 range of hydrate saturations, several hydrate morphologies and confinement stress. These data  
80 have also been used in the calibration of previous mechanical models developed for MHBS  
81 [e.g., 39, 40, 48-50], which give us the opportunity to compare and validate our results. The  
82 model performance is found satisfactory over a wide range of test conditions and evidence the  
83 capability of the Hydrate-CASM model at capturing both the trend and magnitude of the stress-  
84 strain and the volumetric response of synthetic MHBS. In addition, the good matching of our  
85 results with the outputs obtained from previous mechanical models for MHBS evidences that  
86 the experimental data examined in this paper can be simply reproduced by (i) considering the  
87 mechanical effect of the reduction of sediment available void volume due to pore invasion with  
88 hydrate and (ii) modifying the sediment elastic stiffness according to hydrate saturation. Our  
89 results also show that accounting for the different initial void ratios of the set of host specimens  
90 used to produce cementing and pore-filling MHBS allows capturing the experimental data  
91 without using any empirical parameters related to the hydrate morphology.

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## 2. CASM MODEL

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95 The Hydrate-CASM extends the formulation of the constitutive model CASM developed by  
96 Yu [46]. The CASM model is selected here because of its simplicity and flexibility in  
97 describing the shape of the yield surface as well as its proven ability to predict the mechanical  
98 behavior of sand, the most likely target for the commercial exploitation of hydrates [51]. The  
99 critical state model CASM is formulated in terms of the state parameter [52] and the spacing  
100 ratio concept, and uses a non-associated flow rule, which is particularly suitable to simulate the  
101 behavior of granular sediments like those examined in this paper [53, 54]. All the parameters  
102 used in the formulation are listed and defined in Table 2.

103

## 104 2.1. State parameter concept

105

106 The state parameter (Eq. 1) is defined in the  $v - \ln(p')$  space as the vertical distance between  
107 the void ratio at the current state and that at the critical state for a given mean effective stress  
108 (Figure 1a):

$$109 \xi = v + \lambda \ln(p') - \Gamma \quad (1)$$

110

111 The magnitude and sign of this parameter play a key role in understanding the densification  
112 mechanism introduced in this paper. The state parameter adopts positive values when the  
113 sediment void ratio is located above the critical state line (CSL) (as in loose sand; Figure 1a),  
114 and negative ones when located below it (as in dense sand; Figure 1c). Sediments with a  
115 positive value of  $\xi$  and subjected to triaxial shear tend to show hardening on the  $p' - q$  stress  
116 space and contractancy as volumetric response (Figure 1b). Instead, sediments with a negative  
117 value of  $\xi$  show a distinctive peak in the deviatoric stress followed by softening before the  
118 critical state is achieved, and dilatancy dominates its volumetric response (Figure 1d).

119

## 120 2.2. CASM yield function

121

122 A total of seven model parameters are required to define the original CASM formulation. Five  
123 of which ( $\lambda, M, \Gamma, \kappa$  and  $\nu$ ), are the same as those in the Cam Clay model [55,56], and the two  
124 additional parameters, denoted by  $n$  and  $r$ , are used to specify the geometrical properties of the  
125 yield function. For a general stress state, the CASM yield function is expressed as:

$$126 f = \left( \frac{q}{Mp'} \right)^n + \frac{1}{\ln(r)} \ln \left( \frac{p'}{p'_0} \right) \quad (2)$$

127 Where  $n$  governs the shape of the yield surface and  $r$  controls its intersection with the critical  
128 state line. Particular combinations of  $n$  and  $r$  allow the intersection between the critical state  
129 line and the yield surface to not necessarily occur at the maximum deviatoric stress (Figure 2)  
130 as happens in Cam-Clay type models. This allows the CASM model to predict local peaks in  
131 the deviatoric stress on the left side of the critical state condition, feature that is widely observed  
132 in geotechnical testing of sand [57, 58]. Certain values of  $n$  and  $r$  can also recover the yield  
133 surface function of the standard and modified Cam-clay models [46].

134

135 Within the yield surface, the behavior is assumed isotropic and elastic, with the elastic  
136 volumetric stress-strain relationship governed by the bulk modulus  $K$  (Eq. 3a) and the elastic  
137 shear by the shear modulus  $G$  (Eq. 3b):

$$138 \quad K = \frac{(1+e)p'}{\kappa} \quad (3a)$$

$$139 \quad G = \frac{3K(1-2\nu)}{2(1+\nu)} \quad (3b)$$

### 140 2.3. Stress-dilatancy relation and plastic potential

141

142 The CASM model uses a non-associated flow rule that follows the stress-dilatancy law  
143 proposed by Rowe [59], which has been applied with success at describing the deformation of  
144 sands and granular materials [46], as well as to simulate the response of MHBS [32, 33].:

$$145 \quad \frac{d\varepsilon_v^p}{d\varepsilon_q^p} = \frac{9(M-\eta)}{9+3M-2M\eta} \quad (4)$$

146 By integrating equation (4), the CASM plastic potential function is obtained as:

$$147 \quad g = 3M \ln\left(\frac{p'}{\varphi}\right) + (3 + 2M) \ln\left(\frac{2q}{p'} + 3\right) + (M - 3) \ln\left(3 - \frac{q}{p'}\right) \quad (5)$$

148 Whose expression does not depend on the hardening parameters and where  $\varphi$  is a size  
149 parameter controlling the size of the plastic potential which passes through the current stress  
150 state  $(p' - q)$ .

### 151 2.4. Hardening parameters

152

153 Similar to Cam-clay type models, the CASM model assumes isotropic changes in the isotropic  
154 yield stress controlled by the incremental plastic volumetric deformation, so that:

$$155 \quad dp'_0 = \frac{(1+e)p'_0}{\lambda-\kappa} d\varepsilon_v^p \quad (6)$$

156

## 3. HYDRATE-CASM FORMULATION

157

158

159 The Hydrate-CASM extends the formulation of the CASM model [46] by implementing the  
 160 subloading surface model [47] and introducing the densification mechanism. We note that  
 161 material parameters  $e$ ,  $v$ ,  $p'_0$  and  $\kappa$  presented in equations 1 to 5 read as  $e_{ah}$ ,  $v_h$ ,  $p'_{0h}$  and  $\kappa_h$   
 162 in the presence of hydrate within the sediment.

163

### 164 *3.1. Hydrate-CASM subloading function*

165

166 It is widely recognized that plastic strains can develop for stress states inside the yield surface;  
 167 its interior is not a purely elastic domain. This feature results in a smooth transition between  
 168 the elastic and the plastic response of soils [60, 61]. González [62] shows that the CASM yield  
 169 function reproduces well the residual soil strength, but generally over-estimates the elastic  
 170 strains and predicts unrealistic sharp transitions between the elastic and elastoplastic states. The  
 171 subloading surface concept [47] is implemented in the present formulation to account for pre-  
 172 yield plasticity that allows capturing a smoother transition between elastic and plastic behavior,  
 173 and a more accurate volumetric response of MHBS. This model assumes the existence of a  
 174 subloading surface that expands/contracts inside the general yield surface keeping its same  
 175 shape. The Hydrate-CASM subloading function is derived from equation 2 as:

$$176 \quad f = \left(\frac{q}{Mp'}\right)^n + \frac{1}{\ln(r)} \ln\left(\frac{p'}{Rp'_{0h}}\right) \quad (7)$$

177 Where  $R$  controls the size of the subloading surface (Table 2) and recovers the original CASM  
 178 yield function for values equal to 1. The evolution of  $R$  is controlled by the norm of the  
 179 incremental plastic strain vector and the subloading parameter ( $u$ ):

$$180 \quad dR = -u \ln R |d\boldsymbol{\varepsilon}^p| \quad (8)$$

#### 181 *3.1.1. Plastic strain*

182

183 The constitutive equation that characterizes an elasto-plastic material can be expressed as the  
 184 following stress-strain relationship:

185

$$186 \quad d\boldsymbol{\sigma}' = \mathbf{D}^e d\boldsymbol{\varepsilon}^e = \mathbf{D}^e (d\boldsymbol{\varepsilon} - d\boldsymbol{\varepsilon}^p) \quad (9a)$$

187

188 With:

189

$$190 \quad \mathbf{D}^e = \begin{bmatrix} K + \frac{4}{3}G & K - \frac{2}{3}G & K - \frac{2}{3}G & 0 & 0 & 0 \\ K - \frac{2}{3}G & K + \frac{4}{3}G & K - \frac{2}{3}G & 0 & 0 & 0 \\ K - \frac{2}{3}G & K - \frac{2}{3}G & K + \frac{4}{3}G & 0 & 0 & 0 \\ 0 & 0 & 0 & G & 0 & 0 \\ 0 & 0 & 0 & 0 & G & 0 \\ 0 & 0 & 0 & 0 & 0 & G \end{bmatrix} \quad (9b)$$

191

$$192 \quad d\boldsymbol{\varepsilon}^p = d\lambda^p \frac{\partial g}{\partial \boldsymbol{\sigma}'} \quad (9c)$$

193

194 The elastoplastic regime is reached when the stress state lies on the Hydrate-CASM yield  
 195 surface. For the stress state to remain on it at any plastic loading, the consistency condition  
 196 must be satisfied:

197

$$198 \quad df(\boldsymbol{\sigma}', \boldsymbol{\chi}) = 0 \quad (10)$$

199 By linearizing the consistency condition,  $df$  can be rewritten as:

$$200 \quad df = \left(\frac{\partial f}{\partial \boldsymbol{\sigma}'}\right)^T d\boldsymbol{\sigma}' + \left(\frac{\partial f}{\partial \boldsymbol{\chi}}\right)^T d\boldsymbol{\chi} = 0 \quad (11a)$$

201 with:

$$202 \quad \frac{\partial f}{\partial \boldsymbol{\sigma}'} = \frac{\partial f}{\partial p'} \frac{\partial p'}{\partial \boldsymbol{\sigma}'} + \frac{\partial f}{\partial q} \frac{\partial q}{\partial \boldsymbol{\sigma}'} \quad (11b)$$

203

$$204 \quad \frac{\partial f}{\partial \boldsymbol{\chi}} = \left(\frac{\partial f}{\partial p'_{oh}} + \frac{\partial f}{\partial R}\right) \quad (11c)$$

205 By solving equations 9c and 10 the plastic multiplier is classically obtained as:

$$206 \quad d\lambda^p = \frac{\left(\frac{\partial f}{\partial \boldsymbol{\sigma}'}\right)^T \mathbf{D}^e d\boldsymbol{\varepsilon}}{H + \left(\frac{\partial f}{\partial \boldsymbol{\sigma}'}\right)^T \mathbf{D}^e \frac{\partial g}{\partial \boldsymbol{\sigma}'}} \quad (12a)$$

207 where:

$$208 \quad H = - \left( \frac{\partial f}{\partial p'_{oh}} \frac{\partial p'_{oh}}{\partial d\varepsilon_v^p} + \frac{\partial f}{\partial R} \frac{\partial R}{\partial |d\varepsilon^p|} \right) \delta^T \frac{\partial g}{\partial \boldsymbol{\sigma}'} \quad (12b)$$

$$209 \quad \delta^T = \{1,1,1,0,0,0\} \quad (12c)$$

210

### 211 3.2. *Densification mechanism*

212

213 In nature, variations in the sediment void volume may result from two competing and  
214 interdependent processes: (i) mineral precipitation or dissolution (which compares here to  
215 hydrate formation and dissociation, respectively) and (ii) mechanical compaction or dilation  
216 under pressure [63]. In particular, mineral precipitation in pores reduces the sediment available  
217 void volume without experiencing mechanical compaction [64 and 65] and has a significant  
218 effect on its hydraulic and mechanical properties [e.g., 63, 66].

219

220 Figure 3 examines qualitatively the effect of sediment density or void ratio on the magnitude  
221 of  $\xi$  and the corresponding mechanical behavior of the sediment under triaxial shear. For a  
222 reference sediment with positive  $\xi$  (grey cross in Figure 3a and 3b), an increase in density or a  
223 reduction of the void ratio reduces the vertical distance between the current state and the CSL  
224 (black cross in Figure 3b). Thus, during shear, the model predicts less hardening and  
225 contractancy than that observed on the reference sediment (Figure 3c). For a reference sediment  
226 with negative  $\xi$  (grey cross in Figure 3d and 3e), an increase in density increases the distance  
227 of the current state from the CSL (black cross in Figure 3e), and consequently, during shear,  
228 the model predicts a higher peak strength and greater dilatancy than that observed on the  
229 reference sediment (Figure 3f).

230

231 Figure 3 shows that variations in  $\xi$  related to an increase in sediment density produce a similar  
232 mechanical response than those observed in sediments with increasing hydrate saturation (i.e.,  
233 greater strength and dilatancy, or less contractancy, compared to the sediment without hydrate).  
234 Thus, we suggest that the occurrence of hydrate as a solid phase invading the voids of the  
235 hosting sediment may have a similar mechanical effect than the increase of the host sediment  
236 density. Alike Gupta et. al. [67], the Hydrate-CASM formulation conceptually divides the  
237 sediment void-space into potential void volume ( $V_v$ ) and available void volume ( $V_a$ ) (Figure 4).  
238 The potential void volume is the space between the mineral grains of the sediment and includes  
239 the available void volume for fluid flow and storage and the hydrate volume.

240

241



242 To introduce the densification effect that pore invasion with hydrate has on the mechanical  
 243 response of the sediment; the Hydrate-CASM uses the available void ratio left after hydrate  
 244 formation (Eq.13) to derive the mechanical properties of the sediment.

245

$$246 \quad e_{a_h} = e(1 - S_h) = e - e_h \quad (13)$$

247 From where, variations in  $\xi$  with hydrate saturation can be derived as:

$$248 \quad d\xi = de_h \quad (14)$$

249 In addition to the reduction of sediment available void ratio, the presence of hydrate also  
 250 enhances the sediment stiffness [16, 22, 23]. We represent the stiffening effect of hydrate on  
 251 the elastic response of the sediment by the following explicit dependency between  $\kappa$  and  $S_h$ :

$$252 \quad \kappa_h = \kappa \kappa_{rf} \quad (15)$$

253 With:

$$254 \quad \kappa_{rf} = \begin{cases} 1, & S_h = 0 \\ 3S_h^2 - 2.68S_h + 0.9934, & 0 < S_h \leq 0.42 \\ 0.397, & S_h > 0.42 \end{cases} \quad (16)$$

255 Equation 16 is obtained empirically by calibrating the experimental data of three synthetic  
 256 sediments with hydrate saturations ranging from 24.2% to 53.1% (data examined in section  
 257 4.2). This empirical relation needs validation for other sediments and hydrate saturations  
 258 outside the range used for its determination.

259 The decrease of  $\kappa$  in MHBS has been recently observed in experimental high-pressure  
 260 oedometer tests [68]. In our formulation the use of  $\kappa_{rf}$  compensates for spurious changes of  $K$   
 261 (Eq. 3a) when reducing the sediment available void ratio with increasing  $S_h$ . If neglecting the  
 262 hydrate-related stiffening effect suggested in Eq.15, the Hydrate-CASM is still capable of  
 263 reproducing a close solution to the experimental results (purple line in Figure 5b). However,  
 264 the use of  $\kappa_h$  adopted in this work leads to a better fit of the elastic response and the peak  
 265 strength of synthetic hydrate-bearing sediments subjected to triaxial shear (red line in Figure  
 266 5b).

267 As a result of both the decrease of the host sediment available void ratio and the increase of its  
 268 stiffness during hydrate formation, a greater isotropic yield stress can be deduced graphically  
 269 in the  $v - \ln(p')$  space by projecting  $e_{ah}$  on the normal consolidation line (NCL) of the host  
 270 sediment following the  $\kappa_h$  slope (Figure 5a), so that:

$$271 \quad p'_{0h} = \exp\left(\frac{e_h}{\lambda - \kappa_h}\right) p'_0 \left(\frac{\lambda - \kappa}{\lambda - \kappa_h}\right) \quad (17a)$$

272 Where changes in  $p'_{0h}$  are computed through  $dp'_{0h}$ , which reads:

$$273 \quad dp'_{0h} = \frac{(1 + e_{ah})p'_0}{\lambda - \kappa} d\varepsilon_v^p \quad (17b)$$

274

### 275 *3.2.1. MHBS critical state*

276 To evaluate the influence of the densification mechanism due to hydrate formation in the  
 277 critical state of the sediment, Figure 6b relates the potential void ratio of the host sediment ( $e$ )  
 278 with the isotropic yield stress predicted after hydrate formation ( $p'_{0h}$ ).

279 Figure 6a shows the procedure to obtain the isotropic yield stress of the MHBS ( $p'_{0h}$ ), for which  
 280 the sediment with hydrate is considered mechanically denser ( $e_{ah} < e$ ) and stiffer ( $\kappa_h < \kappa$ )  
 281 than the corresponding host sediment. When relating  $p'_{0h}$  with the potential void ratio of the  
 282 sediment ( $e$ ), both the NCL and CSL move to the right in the  $v - \ln(p')$  space (Figure 6b).  
 283 Thus, for a given  $S_h$  the model predicts a normal consolidation line  $NCL_h$  that is parallel to that  
 284 for the host sediment (NCL) and that keeps a vertical distance from the  $CSL_h$  equal to  $\xi_r$  (Table  
 285 2).

286

### 287 *3.2.2. Hydrate dissociation phenomena*

288

289 Several experimental studies [69-74], and field observations [7-9, 13, 14] have demonstrated  
 290 the impact of hydrate dissociation in the mechanical properties of MHBS. Hydrate dissociation  
 291 occurs when the P-T and salinity conditions of the system are outside the hydrate stability zone.  
 292 In the case of hydrate dissociation, the available void ratio of the sediment increases  
 293 proportionally to the volume of hydrate dissociated, which in turn increases the sediment  
 294 permeability and reduces its stiffness and strength [22, 75]. Consequently, stress changes and  
 295 mechanical deformation might be expected during specific conditions of hydrate dissociation.

296 This aspect is integrated in the model since equations 13 to 17b predict an increase in both  $e_{ah}$   
297 and  $\kappa_h$ , as well as a decrease in  $p'_{0h}$  with decreasing  $S_h$ .

298

299 Figures 7 and 8 examine qualitatively the performance of the model in two different scenarios  
300 of thermal-induced hydrate dissociation under constant effective stress.

301 Figure 7 shows the ability of the model at predicting sediment collapse induced by hydrate  
302 dissociation after isotropic consolidation. Upon hydrate dissociation, the sediment is assumed  
303 to recover the mechanical properties of the host sediment (i.e., NCL and CSL). Then, and as  
304 observed by Yoneda's et al. [68] observations, if after the hydrate dissociation the  $v -$   
305  $\ln(p')$  state of the sediment is located in a mechanically inadmissible stress state (point 4 in  
306 Figure 7c) the model can predict sediment collapse until reaching a normally consolidated state  
307 (point 5 in Figure 7c).

308

309 Figure 8 examines the deformation properties of a hydrate-free specimen and a dissociated  
310 MHBS during triaxial shear. Initially, both sediments are isotropically consolidated up to  $p'_{iso}$   
311 (Figures 8a and 8c). After consolidation, the MHBS is subjected to dissociation under constant  
312 effective stress (point 3, Figure 8d), so that the mechanical properties of the host sediment are  
313 recovered (i.e., NCL and CSL, Figure 8d). Then, both sediments are sheared under drained  
314 conditions. In agreement with experimental observations in synthetic MHBS subjected to  
315 dissociation after isotropic consolidation [75], our model predicts a lower failure strength for  
316 the MHBS after dissociation than that observed in the host sediment during shear (Figure 8e).  
317

318

#### 4. HYDRATE-CASM PERFORMANCE

319

320 Triaxial tests at constant hydrate saturation provide very useful information to understand the  
321 influence of hydrate saturation on the mechanical behavior of MHBS. Two sets of stress-strain  
322 data from published triaxial tests are used here to evaluate the model performance. The selected  
323 experimental data report the mechanical behavior of synthetic MHBS subjected to drained  
324 triaxial shear at different confining effective stress, hydrate morphology and saturation. This  
325 data have been widely used to calibrate previous mechanical models developed for MHBS,  
326 which allows us to compare the model results and validate our formulation.

327

328 *4.1. Modeling of Masui's et al. (2005) experimental tests*

329

330 Masui et al. [19] conducted several triaxial tests on synthetic MHBS with different hydrate  
331 saturation and morphologies. Toyoura sand specimens with slightly different void ratios (Table  
332 3) were used as host sediments for the synthetic formation of hydrate with cementing and pore-  
333 filling morphologies. Prior to forming hydrate, the host sediments were isotropically  
334 consolidated up to 1 MPa of confining effective stress. Then, the ice-seed method and the  
335 partial water saturation method were employed to produce hydrates with dominant pore-filling  
336 and cementing morphologies, respectively. After hydrate formation, the hydrate-bearing sand  
337 specimens were sheared at a constant rate of  $0.1 \text{ \% min}^{-1}$  in drained conditions.

338

339 The set of critical state parameters characterizing the behavior of pure Toyoura sand (i.e.,  
340 hydrate-free sediment) (Table 4) have been calibrated here using the stress-strain curve and the  
341 volumetric response of the host specimen used for the synthetic formation of cementing hydrate  
342 ( $S_h=0\%$  in Figure 9c). For the calibration process, values adopted in previous publications that  
343 also model the mechanical response of Toyoura sand have been used as a reference [e.g., 41,  
344 45, 49]. In addition, the different void ratios of 0.6 and 0.75 reported for the cementing and  
345 pore-filling specimens respectively, have also been considered in the simulations (Table 4).

346

347 Figure 9 shows the model results for Masui's et al. [19] triaxial tests. Overall, our results are  
348 satisfactory if one keeps in mind the simplicity of the model formulation in terms of the number  
349 of input parameters required. The Hydrate-CASM successfully captures the trend and  
350 magnitude of the mechanical response of MHBS subjected to shear, showing an increase in  
351 stiffness, shear strength, and dilatancy with increasing  $S_h$  (Figures 9c to 9f). The model outputs  
352 fit particularly well the volumetric response of the cementing specimens (Figure 9e) as well as  
353 the rate of increase observed in the peak strength with  $S_h$  (Figure 9f). However, they  
354 underestimate the maximum deviatoric stress of the cementing specimen with  $S_h=55.1\%$   
355 (Figure 9c) and slightly overestimate the maximum deviatoric stress of the pore-filling  
356 specimen with  $S_h=26.4\%$  (Figure 9d) and the volumetric response of the pore-filling sediment  
357 with  $S_h=40.9\%$  (Figure 9e).

358

359 Previous mechanical models for MHBS that also modelled Masui's et al. [19] experimental  
360 data [e.g., 39, 41, 50] assume that the differences in strength and dilatancy observed between

361 cementing and pore-filling specimens for a given hydrate saturation are controlled by hydrate  
362 morphology. However, Masui et al. [19] state that if the pore hydrate saturation is the same in  
363 both types of specimens (e.g.,  $S_h \approx 40\%$  in Figures 9c and 9d), shear strength becomes higher  
364 for the specimen with lower void ratio. The similarity between the results from previous models  
365 and those obtained with the Hydrate-CASM (Figure 10), which does not consider mechanical  
366 contributions related to hydrate morphology, suggests that the different mechanical behavior  
367 between cementing and pore-filling specimens can be alternatively reproduced considering the  
368 different void ratios reported for each set of the host specimens (Table 3).

369

#### 370 *4.2. Modeling of Hyodo's et al. (2013) experimental tests*

371

372 Hyodo et al. [21] performed a series of triaxial tests to investigate the mechanical properties  
373 and dissociation characteristics of synthetic MHBS. They used an innovative temperature  
374 controlled high-pressure apparatus specially developed to reproduce the in-situ conditions  
375 expected during gas extraction from hydrates. Three sets of triaxial tests conducted at zero or  
376 constant hydrate saturation are used here for the model application. The tests were performed  
377 on Toyoura sand with an initial porosity of about 40% ( $e \approx 0.65$ ), subjected to confining  
378 effective stress of 1, 3 and 5 MPa with different hydrate saturations. The experimental data  
379 from the host sediments (i.e., hydrate-free specimens) are used to calibrate the critical state  
380 parameters of the model (Table 5) and those from the hydrate-bearing sand are used to examine  
381 the model capability at capturing the mechanical effect of  $S_h$ .

382

383 Figure 11 shows the simulation of the experimental tests performed by Hyodo et al. [21]. The  
384 results show the capability of the Hydrate-CASM at capturing changes in the mechanical  
385 response of the host sediment with increasing confining effective stress. For the host sediment  
386 confined at 1 MPa the model predicts a moderate softening after a peak and the volumetric  
387 strain goes from compressive to slightly dilatant ( $S_h=0\%$ , Figure 11a). With increasing  
388 effective stress, the model predicts a gradual transition of this response towards a hardening  
389 and a fully contracting behavior, although the maximum deviatoric stress at 3 and 5 MPa are  
390 slightly underestimated ( $S_h=0\%$ ; Figure 11c and 11e). The results for the hydrate-bearing sand  
391 show, in general, a good agreement with the experimental data, capturing both the trend and  
392 magnitude of the stress-strain and volumetric responses of the sediment (Figure 11a, c and 11e)  
393 and the  $e - \ln(p')$  paths during triaxial shear (Figure 11b, 11d and 11f). However, the model

394 largely overestimated the peak strength for the sediment with  $S_h = 53.7\%$  tested at 3 MPa  
395 (Figure 11c).

396

397 The maximum strength of the sediments examined in this section tends to increase almost  
398 linearly with hydrate saturation. However, the sediment with  $S_h = 53.7\%$  does not follow this  
399 trend (Figure 12a). Hyodo et al. [21] estimated the hydrate saturation within the sediment based  
400 on the stoichiometry of the hydrate formation reaction and assuming that all the methane gas  
401 injected converted into hydrate. Several studies have proposed that hydrate and gas can coexist  
402 under hydrate stability conditions [76-78]. In particular, Sahoo et al. [79] show experimental  
403 evidence in which hydrate formation stops with up to 13% of gas still on the sediment under  
404 favorable pressure, temperature and salinity conditions. Accordingly, we hypothesize that is  
405 possible that part of the gas injected into the specimen with  $S_h=53.7\%$  could not form hydrate  
406 and consequently, the saturation reported could have been slightly overestimated. For  
407 comparison purposes, the same test was modelled considering  $S_h= 24.2\%$ , which is a more  
408 consistent value within the linear  $q_{max} - S_h$  trend observed for the rest of the experimental  
409 data (Figure 12a). Considering  $S_h= 24.2\%$ , the Hydrate-CASM reproduces closely the  
410 deviatoric stress-axial strain relationship reported experimentally (Figure 12b).

411

412 The results presented in this section have been validated against the outputs from three other  
413 mechanical models for MHBS [41, 48, 49] (Figure 13). The comparison is satisfactory and  
414 shows that, despite the simplicity of the densification mechanism, the Hydrate-CASM  
415 performs similarly to models that require more than one hydrate-related empirical parameters  
416 in their formulation.

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## 5. CONCLUSIONS

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422 The Hydrate-CASM is a new elastoplastic constitutive model developed to simulate the  
423 mechanical behavior of MHBS. This model extends the formulation of the CASM model by  
424 implementing the subloading surface model and introducing the densification mechanism.  
425 Alternatively to bonding or cementing models, the Hydrate-CASM suggests that the greater  
426 strength and dilatancy observed in MHBS can be explained by the densification and stiffening  
427 effects that pore invasion with hydrate has on the mechanical properties of the sediment. The

428 densification mechanism attributes hydrate-related changes in the host sediment available void  
429 ratio, swelling line slope and isotropic yield stress to sediment stress-strain changes. Moreover,  
430 the flexibility in the shape of the Hydrate-CASM yield function and the use of a non-associated  
431 flow rule make our formulation particularly suitable for modelling the behavior of sands, the  
432 most likely target deposit for commercial exploitation of hydrates.

433

434 Compared to previous models for MHBS, our formulation reduces to one the number of  
435 empirical hydrate-dependent parameters required to reasonably capture the mechanical  
436 behavior of MHBS. The Hydrate-CASM only requires an empirical hydrate-dependent  
437 parameter to express changes in the sediment swelling line slope with hydrate saturation.  
438 Reducing to one the number of these parameters is an important advance in mechanical  
439 constitutive modeling of MHBS (i) because obtaining them through laboratory tests is  
440 challenging, especially if their physical meaning is not well understood, and (ii) because eases  
441 the application of the Hydrate-CASM model to a wide range of experimental test conditions.

442

443 Robust and well-described published experimental tests have been chosen to calibrate the  
444 Hydrate-CASM capabilities at modelling the mechanical behavior of MHBS during triaxial  
445 shear. These tests cover the most relevant conditions related to MHBS behavior, including a  
446 wide range of hydrate saturations, several hydrate morphologies and confinement stress. In  
447 addition, they have been previously used to calibrate other mechanical models developed for  
448 MHBS, which allowed us to compare and validate our results. Our simulations evidence the  
449 ability of the Hydrate-CASM to predict both stress-strain and the volumetric response of  
450 synthetic MHBS subjected to triaxial shear and suggest that quantifying the void ratio and the  
451 mechanical response of the host sediment is key to isolate hydrate-related mechanical  
452 contributions.

453

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458

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655 **Table 1:** Notable mechanical models for MHBS considering hydrate-bonding effect.

Model reference	Hydrate-bonding modelling strategy
Klar et al. [35]; Jung et al. [36]; Pinkert and Grozic, [37]; Pinkert et al. [38]	Additional cohesion constituent in the failure criteria
Uchida et al. [39]; Sánchez and Gai [40]; Sánchez et al. [41].	Enlargement of the yield surface by cohesion and dilatation
Sultan and Garziglia et al. [42]	Impediment of the normal consolidation of the sediment and enlargement of the yield surface
Sánchez and Gai [40]; Sánchez et al. [41].De La Fuente et al. [43]	Stress-strain partition between hydrate and matrix in a bonding damage framework (BDM)
Jiang et al. [44]	Attribution of physical bonding properties in discrete element methods (DEM)
Lin et al. [45]	Expansion of the failure envelope in a spatially mobilized plane (SMP) model

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668 **Table 2.** CASM and Hydrate-CASM parameters. Subscript  $h$  refers to hydrate-bearing  
669 sediment properties and bold symbols denote tensors. Note that  $e_{ah}$ ,  $v_h$ ,  $p_{0h}$  and  $\kappa_h$  recover  
670 the hydrate-free parameters  $e$ ,  $v$ ,  $p_0$  and  $\kappa$  when  $S_h=0$ .

Model parameters	Description
<b>Stress</b>	$P_p$ Pore pressure
	$\boldsymbol{\sigma}$ Cauchy total stress tensor
	$\mathbf{I}$ Identity matrix
	$\boldsymbol{\sigma}'$ Cauchy effective stress tensor, $\boldsymbol{\sigma}' = \boldsymbol{\sigma} - P_p \mathbf{I}$
	$\boldsymbol{\sigma}_c$ Confining total stress
	$\boldsymbol{\sigma}'_c$ Confining effective stress, $\boldsymbol{\sigma}'_c = \boldsymbol{\sigma}_c - P_p$
	$p$ Mean stress, $p = 1/3 (\sigma_1 + \sigma_2 + \sigma_3)$
	$q$ Deviatoric stress, $q = \sigma_1 - \sigma_3$
	$p'$ Mean effective stress, $p' = p - P_p$
	$\eta$ Stress ratio $\eta = q/p'$
<b>Strain</b>	$\boldsymbol{\varepsilon}$ Total infinitesimal strain tensor
	$\boldsymbol{\varepsilon}^e$ Elastic strain tensor
	$ d\varepsilon^p $ Norm of the incremental plastic strain vector
	$\varepsilon_v^p$ Plastic volumetric strain, $\varepsilon_v^p = \varepsilon_1^p + \varepsilon_2^p + \varepsilon_3^p$
	$\varepsilon_q^p$ Plastic deviatoric strain, $\varepsilon_q^p = \frac{2}{3}(\varepsilon_1^p - \varepsilon_3^p)$
<b>Volumetric ratios</b>	$V_t$ Total volume
	$V_s$ Volume of mineral grains
	$V_h$ Volume of hydrate
	$V_v$ Potential void volume, $V_v = V_t - V_s$
	$V_a$ Available void volume, $V_a = V_v - V_h$
	$e$ Void ratio of the host sediment, $e = V_v/V_s$
	$S_h$ Hydrate saturation, $S_h = V_h/V_v$
	$e_h$ Hydrate ratio, $e_h = V_h/V_s = S_h e$
	$e_{ah}$ Available void ratio of the hydrate-bearing sediment, $e_{ah} = e(1 - S_h)$
	$v$ Specific volume, $v = 1 + e$
$v_h$ Hydrate-CASM equivalent specific volume, $v_h = v - e_h$	
<b>Critical state parameters</b>	$\lambda$ Slope of the normal compression and critical state lines in the $v - \ln(p')$ space
	$M$ Critical state stress ratio: slope of critical state line in the $p' - q$ space
	$p_0$ Isotropic yield stress of the host sediment
	$p_{0h}$ Isotropic yield stress of the hydrate-bearing sediment
	$p'_x$ Mean effective stress at critical state
	$\Gamma$ Specific volume at critical state with $p'$ of 1 KPa
<b>Elastic parameters</b>	$\kappa$ Host sediment swelling (reloading-unloading) line slope
	$\kappa_h$ MHBS swelling (reloading-unloading) line slope
	$\nu$ Poisson's ratio
	$K$ Elastic bulk modulus
	$G$ Elastic shear modulus
	$\mathbf{D}^e$ Elastic stiffness tensor
<b>CASM parameters</b>	$n$ Stress-state coefficient: yield surface shape parameter
	$r$ Spacing ratio, $r = p'_0/p'_x$
	$\xi$ State parameter
	$\xi_r$ Reference state parameter, $\xi_r = (\lambda - \kappa) \ln r$

<b>Subloading parameters</b>	$p_{0_s}$	Isotropic yield stress of the subloading surface
	$R$	Subloading surface ratio, $R = p_{0_s}/p_0$
	$u$	Subloading parameter controlling plastic deformations before yielding
<b>Plastic parameters</b>	$\varphi$	Size parameter
	$\mathcal{X}$	Vector of hardening (2 components: $p_{0_h}$ and $R$ )
	$H$	Hardening modulus
	$\lambda^p$	Plastic multiplier
<b>Empirical parameters</b>	$\kappa_{rf}$	Swelling line reduction factor

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691 **Table 3.** Physical properties of Toyoura sand specimens used in Masui et al. [19] as host  
692 sediment for the synthesis of cementing and pore-filling hydrates.

<b>Host sediment physical properties</b>		
	<b>Cementing specimens</b>	<b>Pore-filling specimens</b>
Diameter/height (mm)	50/100	50/100
Density (g/cm <sup>3</sup> )	1.74–1.92	1.77–1.78
Void ratio	0.57-0.63	0.73-0.75

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715 **Table 4.** Host sediment input parameters for modeling Masui et al. [19] triaxial tests.

<b>Host sediment input parameters</b>									
$e$	$\lambda$	$M$	$p'_0(MPa)$	$\kappa$	$\nu$	$n$	$r$	$p'_{0s}(MPa)$	$u$
<b>Cementing specimens</b>									
0.6	0.22	1.17	12	0.015	0.1	2.5	1.7	3.5	20
<b>Pore-filling specimens</b>									
0.75	0.22	1.17	5.3	0.015	0.1	2.5	1.7	3	20

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737 **Table 5.** Host sediment input parameters for modelling Hyodo's et al. [21] triaxial tests at 1, 3  
 738 and 5 MPa of confining effective stress.

<b>Host sediment input parameters</b>									
$e$	$\lambda$	$M$	$p'_0(MPa)$	$\kappa$	$\nu$	$n$	$r$	$p'_{0s}(MPa)$	$u$
0.65	0.22	1.32	9	0.015	0.1	4	2.5	5.6	50

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