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A model to predict the elastic properties of gas hydrate-bearing sediments

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Abstract: A simple and accurate model for predicting the elastic properties of gas hydrate-bearing sediment (GHBS) is proposed and validated against experimental data. It is developed on the basis of the homogenization theory for multiphase composite. Unlike the classical homogenization techniques those fix a homogenization scheme (*e.g.* self-consistent, Mori-Tanaka, DEM, *etc.*) for a given microstructure, the proposed model considers a flexible homogenization scheme that adapts with the change of the microstructure of the material. The idea is to modify the elastic properties of the reference matrix with respect to the microstructure change of the mixture. Such modification is ensured by adapting the theoretical method to available experimental data. The derived method is proved to be very powerful as it can fit laboratory measured data on gas hydrate-bearing sand formed by different methods including excess gas and excess water methods and log data taken from various sites. Such feature cannot be satisfied by the existent models those consider the hydrate phase as a cement phase or a pore-filling phase. A very good agreement between the proposed method and experimental data offer a simple and accurate ways to predict the saturation of gas hydrate using sonic log data or 3D seismic data.

Keywords: gas hydrate-bearing sediment; elastic properties; homogenization; adaptive scheme

1. Introduction

Gas hydrate-bearing sediment (GHBS) is an energy resource made of low molecular weight gas (methane) hosted by hydrogen-bonded water filling in the porous space between the solid

35 sediment grains (Milkov, 2004; Sloan and Koh, 2007; Waite et al., 2009; Rees et al., 2011).
36 The dissociation of gas hydrate in sediment nearby a borehole yields the change of the in situ
37 pore pressure and stress as well as the modification of the mechanical properties of the GHBS
38 (Li et al., 2011; Le et al., 2019). These phenomena can lead to serious stability problems (Tan
39 et al., 2005; Rutqvist and Moridis, 2007; Freij-Ayoub et al., 2007).

40 The gas hydrate content acts as a lightweight solid content that decreases the hydraulic
41 conductivity and increases the elastic stiffness and the sonic wave velocities of the GHBS
42 comparing to equivalent sediment without gas hydrate (Priest et al., 2005; Johnson et al., 2011).
43 Therefore sonic log data can be used to estimate hydrate saturation (Guerin et al., 1999).

44 A number of models were proposed to predict the elastic and sonic properties of GHBS.
45 Lee et al. (1996) proposed a simple weighted equation to estimate the hydrate saturation using
46 compressional and shear sonic wave velocities (see also Lee and Collett, 2001; Lee, 2002).
47 Carcione and Tinivella (2000) use a Biot-type three-phase model including two solid phases to
48 model the sonic velocities of GHBS. Helgerud et al. (1999) considered an effective medium
49 theory that was previously developed by Dvorkin and Nur (1996) for sandstone to model
50 GHBS. This later considered the contact Hertz-Mindlin model for a granular packing at a
51 critical porosity of about 0.36 to 0.40 and then the Hashin-Shtrikman formulas (Hashin and
52 Shtrikman, 1963) to estimate the effective bulk and shear moduli at porosities higher or lower
53 than the critical porosity. Then the Gassmann's theory (Gassmann, 1951) was employed to
54 relate the dry and saturated moduli. In this model, the hydrate content was regarded by two
55 possibilities: hydrate was associated to the solid phase or the fluid phase. This idea was also
56 considered by Jakobsen et al. (2000) in an anisotropic Self-Consistent (SC) and Differential
57 Effective Medium (DEM) approximation (combined SC-DEM method). For the assumption of
58 hydrate associated to the solid phase, they started by considering the self-consistent scheme for
59 the mixture of solid grains and hydrate, then the multi-step DEM procedure was used to replace
60 a little amount of hydrate by water in each step. The role of hydrate and water were interchanged
61 if hydrate was considered as part of the fluid phase. This model was considered by Chand et al.
62 (2004, 2006) for the computation of the hydrate saturation in GHBS from sonic data. However,
63 when comparing the results predicted by the SC-DEM method with experimental data, we
64 observe that it strongly over-estimates the elastic properties and the sonic velocities of gas
65 hydrate-bearing sediments. Indeed, such method can be used for highly compacted rocks such
66 as shale or sandstone those are buried deeply in the sediments but it is not appropriate for gas
67 hydrate-bearing sediments those are usually located at a shallow buried depth.

68 Recently, a new and powerful homogenization concept was proposed to deal with the
69 effective physical properties of composite containing highly contrast phases (Nguyen et al.,
70 2016). It was a flexible homogenization technique that could smoothly transfer from one
71 homogenization scheme to another by modifying the properties of the reference matrix with
72 respect to the microstructure of the whole medium. It was demonstrated that experimental data
73 can be perfectly fitted by such method.

74 In the present work, the concept proposed by Nguyen et al. (2016) is applied to predict the
75 elastic and sonic properties of GHBS. After a review of experimental observations, the
76 theoretical basis of the homogenization is briefly presented. Afterward, the homogenization
77 scheme is calibrated and validated against experimental data collected from literature. The
78 sensitivity of the model's parameters is analyzed. The robustness of the model to predict the
79 sonic properties of GHBS is finally discussed.

80 **2. Review of experimental observations**

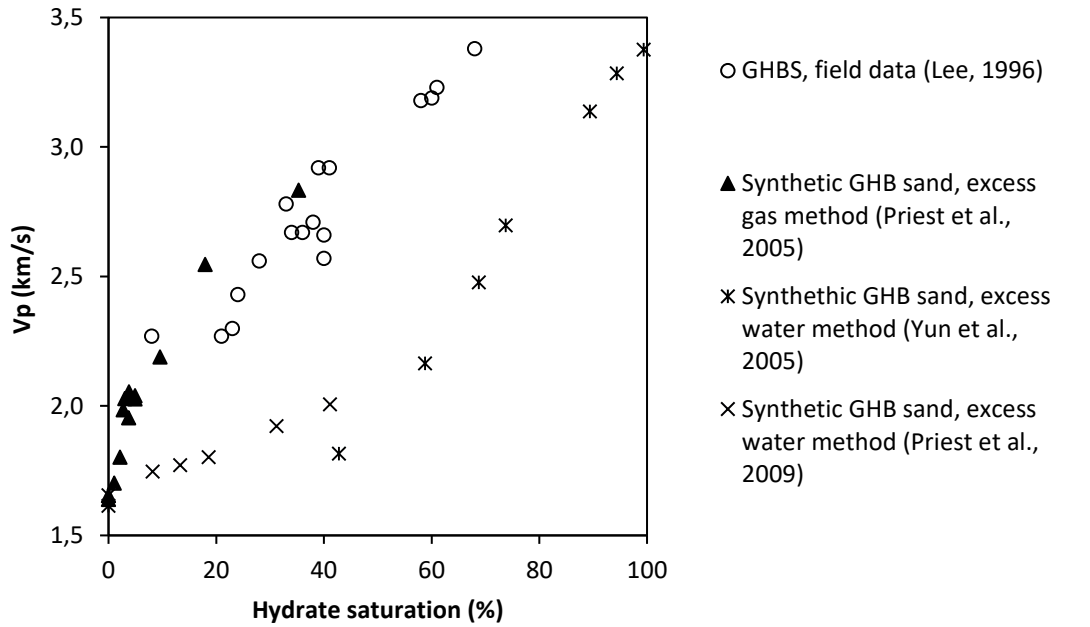
81 Several experimental data on the elastic and sonic properties of GHBS can be found in
82 literature. For example, Priest et al. (2005) measured the compressional and shear wave
83 velocities of gas hydrate-bearing sand samples formed by the technique proposed by Stern et
84 al. (1996). Their measurements were performed at low frequencies and small strain amplitude
85 corresponding to the range of field seismic acquisition. They observed a strong increase of the
86 sonic velocities versus increasing of hydrate saturation. The stress dependence of the sonic
87 velocities, observed for sand packing and sandstone without gas hydrate, was also observed for
88 gas hydrate-bearing sand. Such observation suggests that the role of grain-to-grain contact is
89 also important for GHBS. Yun et al. (2005) measured the compression and shear sonic wave
90 velocities of tetrahydrofuran (THF) gas hydrate-bearing sand. This latter was formed from
91 water saturated sand packing by decreasing the temperature at low confining pressure condition
92 (see also Choi et al, 2014; Sultaniya et al., 2015). They observed that the dependence of the
93 sonic wave velocities on the hydrate saturation of GHBS was a lot less significant than that
94 formed by Priest et al. (2005). It was confirmed by Priest et al. (2009) that the elastic wave
95 velocities of GHBS were strongly affected by its morphology. Indeed, the hydrate-bearing sand
96 formed by “excess gas” method (Priest et al., 2005) created cement gas-hydrate morphology
97 while the “excess water” method considered by Yun et al. (2005) and Priest et al. (2009) created
98 pore-filling gas-hydrate morphology. Hu et al. (2012) created methane hydrate-bearing sand
99 sediment by injecting directly high-pressure methane gas into natural sand saturated with a 300
100 ppm Sodium dodecyl sulfate (SDS) solution. Konno et al. (2015) measured the compressional

101 wave velocity of natural sandy sediments at near in-situ pressures and temperatures and found
102 that the wave velocity correlated well with the hydrate saturation. To summary, there are three
103 main methods to create gas hydrate in laboratory: (i) excess water hydrate formation; (ii) excess
104 gas hydrate formation; and (iii) hydrate formation out of solution. At low saturations, excess
105 gas produces strongly cementing hydrates while excess water and solution techniques produce
106 pore filling hydrates. As the hydrate saturations increase, the hydrate becomes continuously
107 more cementing (see Rydzy, 2015).

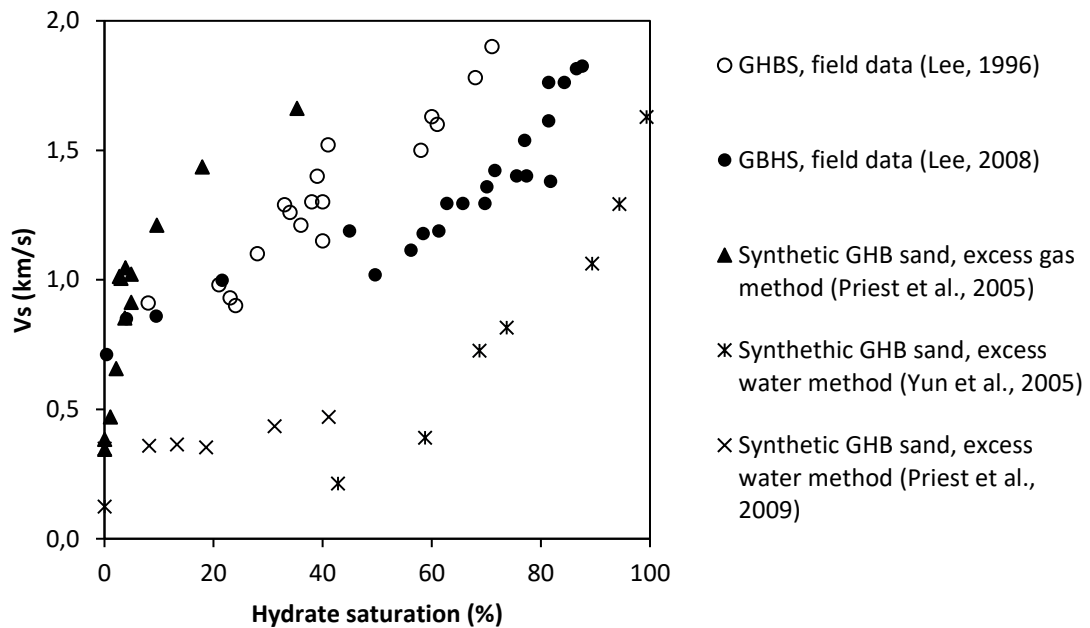
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109 So it is clear that the sonic velocities of GHBS strongly depended on the ways it was formed
110 (Yun et al., 2005; Priest et al., 2005, 2009; Rydzy, 2014) and were different from field data
111 (Lee et al., 1996; Lee and Collett, 2001; Lee, 2008). *Figure 1* and *Figure 2* show the
112 compressional and shear sonic wave velocities (V_s and V_p , respectively), of different gas
113 hydrate-bearing sand formed in laboratory and natural GHBS, versus the hydrate saturation
114 degree as well as a cross-plot between these velocities. Yun et al. (2005) used THF-hydrate
115 instead of methane hydrates because it has similar thermo-mechanical properties to and the
116 same molecular structure (Structure II) as hydrates of most thermogenic gases. In addition, from
117 the micromechanical point of view, the elastic properties of THF and methane hydrates are
118 similar when compared to that of quartz. Data obtained for sandstone and natural sand without
119 gas hydrate (Han et al., 1986; Devorkin and Nur, 1996) are also plotted for comparison. At a
120 low velocity range, corresponding to low hydrate saturation, the shear velocity at a given
121 compressional velocity obtained in laboratory by excess gas method (Priest et al., 2005) is a lot
122 higher than the shear velocity obtained by the excess water method (Yun et al., 2005; Priest et
123 al., 2009). Note that Yun et al. (2005) saturated fine-grained sand specimen by water-THF
124 solutions to form hydrate whereas Priest et al. (2009) injected de-aired water into air-dried fine-
125 grained sand specimen saturated with methane gas. Actually, after Priest et al. (2009), the
126 excess gas method forms gas hydrate at the grain contacts (cementing) while the excess water
127 one allows gas hydrate to form within the pore space between the sand grains (pore-filling). In
128 addition, within the excess water method, at higher hydrate saturation, the hydrate forms at the
129 grain contact (cementing) although hydrate starts to form as pore-filling. For this reason, the
130 results obtained by the excess water method approach those of excess gas method at higher
131 saturation. Some laboratory gas-hydrate forming techniques provide data in between these two
132 limits. For instance, Zhang et al. (2011) formed gas hydrate-bearing sand starting from partially
133 water saturated sand packing and observed that the gas hydrate started to be a pore-filling phase
134 at the beginning and then became the cement content at the end. Of cause other factors such as

135 the stress condition, mineralogy and fluid type must affect the effective elastic and sonic
 136 properties of GHBS. But for a given mineralogy and fluid type at a fixed stress state, it is
 137 obvious that the morphology of the gas-hydrate content must play a crucial role.
 138



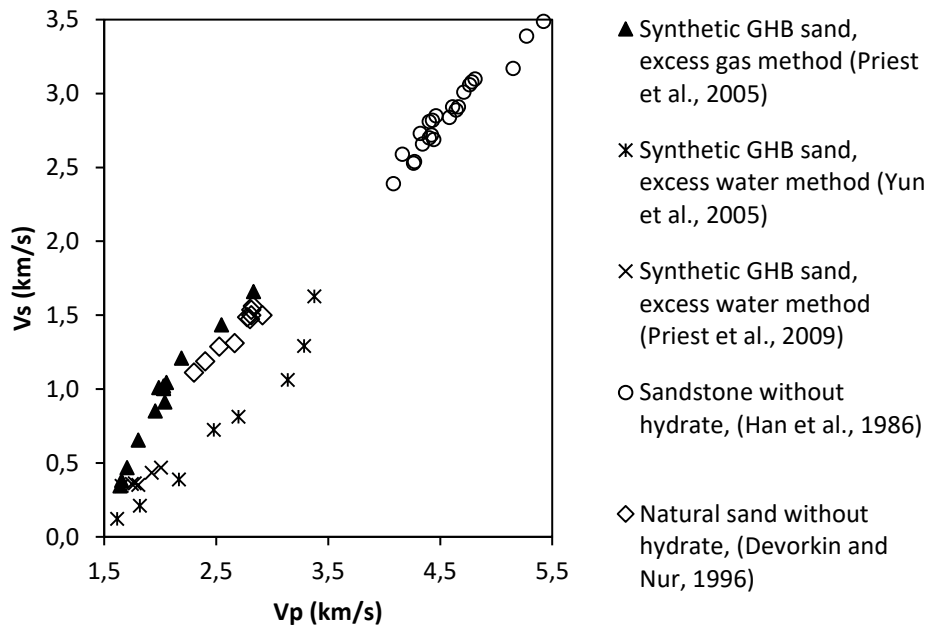
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140

141 *Figure 1: Compressional and shear sonic velocities of GHBS versus hydrate saturation:*
 142 *experimental and field data.*

143



144

145 *Figure 2: Shear sonic velocity (V_s) versus compression sonic velocity (V_p) of gas hydrate-*
 146 *bearing sands and those without gas hydrate.*

147

148 3. Review of the theoretical basis

149 The homogenization theory allows modeling effective elastic (and/or transport) properties
 150 of a heterogeneous medium. It starts by considering an elementary problem of a single
 151 heterogeneity in an infinite medium (dilute problem, Eshelby, 1957). Then Mori-Tanaka theory
 152 (Mori and Tanaka, 1973) allows extending dilute solution to a medium containing many
 153 inclusions those are interacted. However Mori-Tanaka scheme is limited to a matrix-inclusion
 154 system with a low to medium volume fraction of inclusions. Therefore it is not appropriate for
 155 a rock that is not a matrix-inclusion system. In such situation, it is required to define an
 156 equivalent matrix of the medium. For example, the self-consistent scheme considers the
 157 homogenized medium itself as the matrix of the mixture. But this method has also little
 158 application for rocks because it usually over-estimates the elastic properties due to the strong
 159 contrast between the solid and the fluid phases. A more sophisticated definition of the matrix is
 160 required to appropriately modeling elastic (and transport) properties of rock (e.g. Nguyen et al.,
 161 2016)

162 This section provides the basic elements of the homogenization theory for isotropic
 163 composite with a highlight on the flexible homogenization concept previously proposed by

164 Nguyen et al. (2016). The local relationship between the stress tensor $\boldsymbol{\sigma}$ and strain tensor $\boldsymbol{\varepsilon}$ is
 165 defined by the following linear elastic equation:

$$\boldsymbol{\sigma} = \mathbb{C} : \boldsymbol{\varepsilon} \quad (1)$$

166 where \mathbb{C} is the fourth order local stiffness tensor; the column “:” is for a double contraction of
 167 two tensors. The macroscopic behavior of a representative elementary volume (REV) is:

$$\langle \boldsymbol{\sigma} \rangle = \mathbb{C}^{hom} : \langle \boldsymbol{\varepsilon} \rangle \quad (2)$$

168 where the notation $\langle . \rangle$ stands for a volumetric average over the REV; the superscript “hom” is
 169 for a homogenized term. The local and average strains are related via the strain localization
 170 tensor such as:

$$\boldsymbol{\varepsilon} = \mathbb{A} : \langle \boldsymbol{\varepsilon} \rangle \quad (3)$$

171 where \mathbb{A} is the strain localization (or concentration) tensor that is a function of the mechanical
 172 properties, the volume fraction and the shape of the phases as well as the interaction between
 173 them. By combining equations (1) to (3), the effective stiffness tensor \mathbb{C}^{hom} can be related to
 174 the local stiffness tensors as:

$$\mathbb{C}^{hom} = \langle \mathbb{A} : \mathbb{C} \rangle \quad (4)$$

175 According to Mori and Tanaka (1973), the localization tensor \mathbb{A} can be expressed as:

$$\mathbb{A} = \mathbb{A}^{dl} \langle \mathbb{A}^{dl} \rangle^{-1} \quad (5)$$

176 where \mathbb{A}^{dl} is the dilute localization tensor of a matrix-inclusion system that has been well
 177 established by Eshelby (1957):

$$\mathbb{A}^{dl} = [\mathbb{I} + \mathbb{P}_0(\mathbb{C}_{inc} - \mathbb{C}_0)]^{-1} \quad (6)$$

178 where \mathbb{C}_{inc} is the elastic stiffness tensor of the inclusion; \mathbb{C}_0 the elastic stiffness tensor of the
 179 reference matrix; \mathbb{P}_0 the Hill’s tensor that is a function of \mathbb{C}_0 and the shape of the inclusion; \mathbb{I}
 180 the fourth order identity tensor. For the case of isotropic matrix and inclusion, the stiffness
 181 tensors \mathbb{C}_{inc} and \mathbb{C}_0 can be expressed in term of the bulk and shear moduli such as:

$$\mathbb{C}_{inc} = 3k_{inc}\mathbb{J} + 2\mu_{inc}\mathbb{K}; \mathbb{C}_0 = 3k_0\mathbb{J} + 2\mu_0\mathbb{K} \quad (7)$$

182 where \mathbb{J} and \mathbb{K} are the spherical and deviatoric part of the identity tensor \mathbb{I} : $\mathbb{I} = \mathbb{J} + \mathbb{K}$; k_{inc} and
 183 μ_{inc} the bulk and shear moduli of the inclusion; k_0 and μ_0 the bulk and shear moduli of the
 184 reference matrix.

185 The assumption of spherical inclusion is usually considered for sandstone. Such
 186 assumption yields following simple formula of the Hill's tensor (Dormieux et al., 2006):

$$\mathbb{P}_0 = \frac{1}{3(k_0 + a_0)}\mathbb{J} + \frac{1}{2(\mu_0 + b_0)}\mathbb{K} \quad (8)$$

187 where

$$a_0 = \frac{4}{3}\mu_0; b_0 = \frac{\mu_0(9k_0 + 8\mu_0)}{6(k_0 + 2\mu_0)} \quad (9)$$

188 Using the formula (7) for the elastic stiffness tensors of the matrix and inclusion and the
 189 formulas (8) and (9) for the Hill's tensor, equation (6) yields the following formula of an
 190 isotropic dilute localization tensor:

$$\mathbb{A}^{dl} = \frac{k_0 + a_0}{k_{inc} + a_0}\mathbb{J} + \frac{\mu_0 + b_0}{\mu_{inc} + b_0}\mathbb{K} \quad (10)$$

191 Then equation (5) yields:

$$\mathbb{A} = \frac{k_0 + a_0}{k_{inc} + a_0} \left\langle \frac{k_0 + a_0}{k + a_0} \right\rangle^{-1} \mathbb{J} + \frac{\mu_0 + b_0}{\mu_{inc} + b_0} \left\langle \frac{\mu_0 + b_0}{\mu + b_0} \right\rangle^{-1} \mathbb{K} \quad (11)$$

192 where k and μ , which appear inside the average operator $\langle \cdot \rangle$, stand for the bulk and shear moduli
 193 of a component (matrix or inclusion). Note that the terms with index 0 can be put out of the
 194 volumetric average terms, therefore the localization tensor can be simplified to:

$$\mathbb{A} = \frac{1}{k_{inc} + a_0} \left\langle \frac{1}{k + a_0} \right\rangle^{-1} \mathbb{J} + \frac{1}{\mu_{inc} + b_0} \left\langle \frac{1}{\mu + b_0} \right\rangle^{-1} \mathbb{K} \quad (12)$$

195 Finally, the introduction of equation (11) in equation (4) provides the following formula of the
 196 elastic stiffness tensor:

$$\mathbb{C}^{hom} = \left\langle 3k \frac{1}{k + a_0} \right\rangle \left\langle \frac{1}{k + a_0} \right\rangle^{-1} \mathbb{J} + \left\langle 2\mu \frac{1}{\mu + b_0} \right\rangle \left\langle \frac{1}{\mu + b_0} \right\rangle^{-1} \mathbb{K} \quad (13)$$

197 That can be recast in the form:

$$\mathbb{C}^{hom} = 3k_{hom}\mathbb{J} + 2\mu_{hom}\mathbb{K} \quad (14)$$

198 where the homogenized bulk and shear moduli, k_{hom} and μ_{hom} , are (see also Pham et al., 2017):

$$k_{hom} = \left\langle \frac{k}{k + a_0} \right\rangle \left\langle \frac{1}{k + a_0} \right\rangle^{-1} = \left\langle \frac{1}{k + a_0} \right\rangle^{-1} - a_0 \quad (15)$$

199 and

$$\mu_{hom} = \left\langle \frac{\mu}{\mu + b_0} \right\rangle \left\langle \frac{1}{\mu + b_0} \right\rangle^{-1} = \left\langle \frac{1}{\mu + b_0} \right\rangle^{-1} - b_0 \quad (16)$$

200 The choice of the parameters a_0 and b_0 of the reference matrix depends on the
 201 homogenization scheme chosen. For example, the self-consistent scheme corresponds to
 202 computing a_0 and b_0 using equation (9) in which the effective bulk and shear moduli are used
 203 as the bulk and shear moduli of the reference matrix, i.e. $k_0 = k_{hom}$ and $\mu_0 = \mu_{hom}$. The Mori-
 204 Tanaka scheme is usually used for a composite containing a major connected phase that can be
 205 considered as the reference matrix of which the elastic bulk and shear moduli can be used to
 206 compute the parameters a_0 and b_0 . In the multi-step differential effective medium (DEM)
 207 scheme, a_0 and b_0 of a step n are computed from the effective bulk and shear moduli obtained
 208 at the previous step ($n - 1$).

209 The main idea of the homogenization scheme proposed by Nguyen et al. (2016) is to let the
 210 parameters a_0 and b_0 change flexibly according to the change of the microstructure. Such
 211 correlation between the properties of the reference matrix and the microstructure can be
 212 calibrated by inverse analysis regarding available experimental data. Details of this model for
 213 the case of GHBS are presented in subsequent section.

214 4. Application for gas hydrate-bearing sand

215 4.1. The model

216 Considering water saturated gas hydrate-bearing sand that can be modeled by a mixture of
 217 three phases: quartz grains, gas hydrate and water. Applying the formulas (15) and (16), we
 218 have:

$$k_{hom} = \left[\frac{f_q}{k_q + a_0} + \frac{f_h}{k_h + a_0} + \frac{f_w}{k_w + a_0} \right]^{-1} - a_0 \quad (17)$$

219 and

$$\mu_{hom} = \left[\frac{f_q}{\mu_q + b_0} + \frac{f_h}{\mu_h + b_0} + \frac{f_w}{\mu_w + b_0} \right]^{-1} - b_0 \quad (18)$$

220 where f is the volume fraction of a phase; the index q , h and w stand for quartz, gas hydrate
 221 and water, respectively; the bulk and shear moduli of the phases are given in Table 1. The
 222 volume fractions of the phases are related to the porosity and the hydrate saturation by:

$$f_q = 1 - \phi; f_h = \frac{S_h}{100} \phi; f_w = 1 - f_q - f_h \quad (19)$$

223 where ϕ is the initial porosity (porosity at zero hydrate saturation) and S_h (%) is the hydrate
 224 saturation.

225 **Table 1** : *Elastic properties and densities of the phases of GHBS*

	ρ (g/cm ³)	k (GPa)	μ (GPa)	Reference
Phases				
Methal hydrate	0.9	7.9	3.3	Helgerud et al. (1999)
Quartz	2.65	37.8	44.3	Mavko et al. (2009)
Water	1.0	2.3	0.	Mavko et al. (2009)

226

227 In a common homogenization technique, a fixed homogenization scheme is usually
 228 chosen for a composite with a large range of microstructure change. However, it is well-known
 229 that each homogenization scheme can be used for just a certain small range of microstructure
 230 change. For example, the dilute and Mori-Tanaka schemes can be used for small to average
 231 amount of inclusions embedding in a matrix, the self-consistent scheme is appropriate for a
 232 randomly disordered mixture, *etc.* Here we propose to consider an adaptive homogenization
 233 scheme that allows a transition from one homogenization scheme to another. As previously
 234 discussed, the homogenization schemes are differed from each other by the parameters a_0 and
 235 b_0 of the reference matrix. Therefore we propose to adapt these parameters with the
 236 microstructure change; Nguyen et al. (2016) used this method for the effective transport
 237 properties of heterogeneous materials.

238 **4.2. Calibration**

239 Let consider the gas hydrate-bearing sand sample measured by Priest et al. (2005) of
 240 which the hydrate saturation, the volume fraction of water and both compression and shear
 241 sonic velocities were measured. For field application purpose, we consider the sample in water-
 242 saturated state, *i.e.* pore volume is filled by gas hydrate and water. Knowing the volume fraction
 243 of each phase, we compute the bulk density ρ by the following formula:

$$\rho = f_q \rho_q + f_h \rho_h + f_w \rho_w \quad (20)$$

244 Then the sonic velocities V_s and V_p is computed using the homogeneous bulk and shear moduli
 245 as:

$$V_s = \sqrt{\frac{\mu_{hom}}{\rho}}; V_p = \sqrt{\frac{k_{hom} + \frac{4}{3}\mu_{hom}}{\rho}} \quad (21)$$

246 For this calibration step, we calibrate the parameters \mathbf{a}_0 and \mathbf{b}_0 to fit the data provided
 247 by Priest et al. (2005). Results are given in Table 2. It is of interest to note that these data have
 248 been obtained by converting measured data on unsaturated samples to equivalent saturated
 249 samples through the Gassmann's equation. As previously mentioned, the proposed model is
 250 calibrated for saturated samples due to field application purpose. However, the direct
 251 application of the model on unsaturated sample is mentioned later in section 4.4.

252 The following relation between the parameter b_0 and hydrate saturation S_h (%) is
 253 considered to fit the shear velocity (*Figure 3*):

$$b_0 = c_0 \log_{10} S_h, \text{ for } 1\% < S_h < 40\% \quad (22)$$

254 where $c_0 = 2.32$ for this case of gas hydrate sand formed by excess gas method (*Figure 3*). It
 255 is of interest to remark that for hydrate-free section ($S_h \leq 1\%$), rock is uncemented and
 256 unconsolidated then the Reuss approximation $b_0 = a_0 = 0$ (that corresponds to $S_h = 1\%$) is
 257 appropriate (Nur et al., 1991).

258 Then, to fit the compressional velocity (*Figure 4*), we consider a very simple relation:

$$a_0 = \frac{b_0}{2} \quad (23)$$

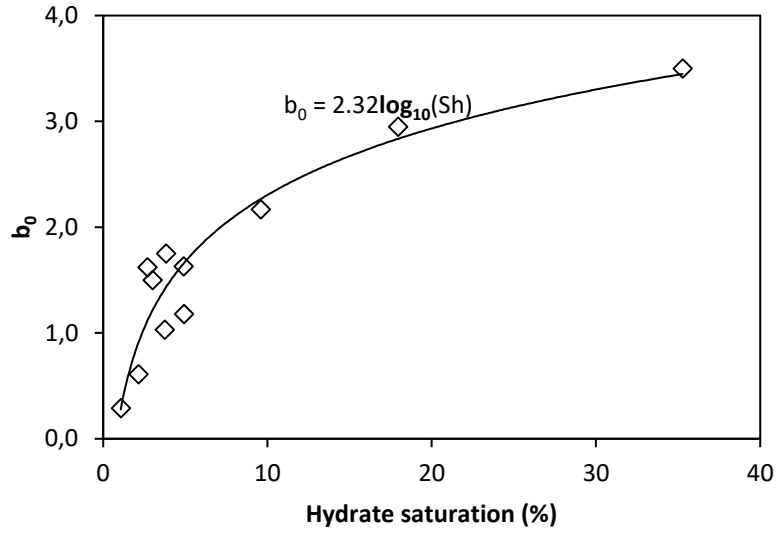
259 To summarize, for each sediment and at a given stress state, the model can predict the
 260 effect of the hydrate saturation on the sonic velocities by using only one fitting parameter c_0 .
 261 The parameters a_0 and b_0 can be then determined by using the equations (22) and (23).

262
 263 **Table 2:** Sonic velocities of GHBS at different phases' volume fractions: a comparison between
 264 modeled and measured data.

S_h (%)	Volume fractions			ρ (g/cm ³)	Modeled			Measured	
	f_w (V/V)	f_h (V/V)	f_s (V/V)		b_0 (GPa)	V_p (km/s)	V_s (km/s)	V_p (km/s)	V_s (km/s)
0.00	0.463	0.000	0.537	1.886	0.20	1.676	0.351	1.639	0.345
0.00	0.413	0.000	0.587	1.969	0.20	1.734	0.380	1.654	0.383
1.07	0.397	0.004	0.599	1.987	0.29	1.808	0.468	1.703	0.470
2.15	0.412	0.009	0.579	1.954	0.61	1.949	0.656	1.802	0.656
3.00	0.409	0.013	0.578	1.953	1.50	2.312	1.006	2.029	1.005
2.70	0.431	0.012	0.557	1.918	1.62	2.303	1.007	1.985	1.012
3.75	0.407	0.016	0.577	1.951	1.03	2.142	0.848	1.955	0.852
3.82	0.429	0.017	0.554	1.912	1.75	2.348	1.045	2.055	1.046

4.91	0.400	0.021	0.579	1.954	1.18	2.215	0.913	2.040	0.913
4.90	0.424	0.022	0.554	1.912	1.63	2.320	1.019	2.028	1.021
9.59	0.392	0.042	0.566	1.930	2.17	2.555	1.209	2.189	1.210
17.95	0.353	0.077	0.570	1.932	2.95	2.845	1.437	2.547	1.435
35.27	0.279	0.152	0.569	1.924	3.50	3.138	1.656	2.833	1.661

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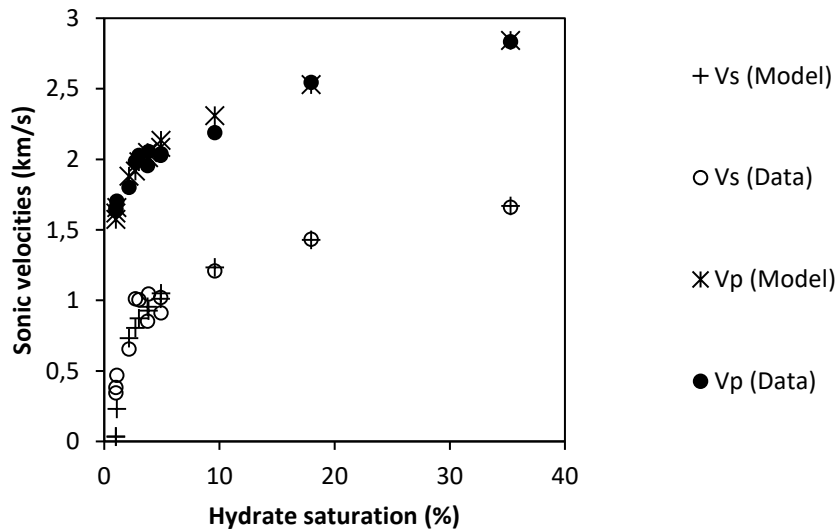


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267 *Figure 3: Dependence of the reference matrix on the microstructure: parameter b_0 versus*
 268 *hydrate saturation S_h .*

269

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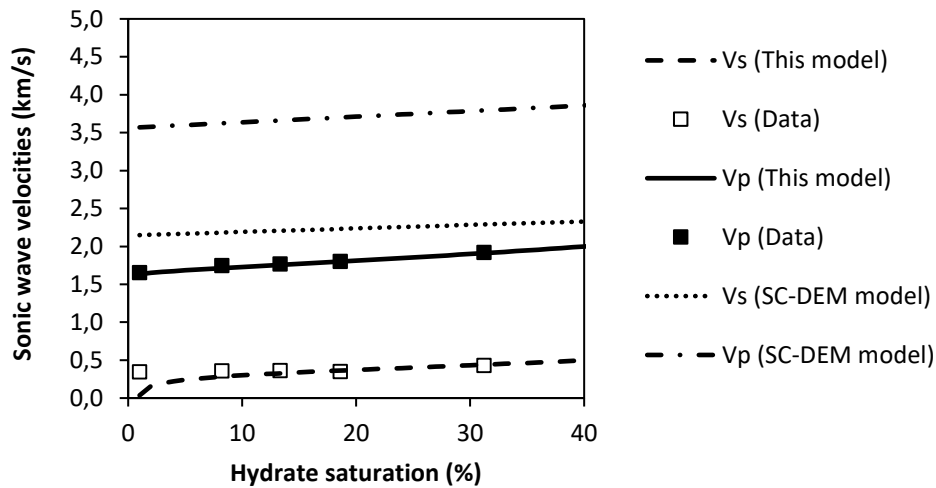


271

272 *Figure 4: Sonic velocity of gas hydrate-bearing sand prepared in laboratory by the excess gas*
 273 *method: Experimental data (Priest et al. 2005) versus prediction by the present*
 274 *model. Parameter: $c_0 = 2.32$.*

275 *Figure 5* shows that a parameter $c_0 = 0.1$ allows a very good fit between the proposed
 276 method and data of gas hydrate-bearing sand formed by excess water method for a range of
 277 hydrate saturation below 40% (Priest et al., 2009). The parameter c_0 obtained for this case is
 278 very small comparing to the previous case of GHBS formed by excess gas method. So the
 279 parameter c_0 can be considered as a good indicator of GHBS formation and morphology.

280 On this figure, the results given by the existent model, that is a combination of the self-
 281 consistent (SC) and differential effective medium (DEM) schemes (Jakobsen et al., 2000;
 282 Hornby et al., 1994) (see also Appendix), are also plotted (called by SC-DEM model) for
 283 comparison. The model presented herein is obviously superior to the existent model. The
 284 existent SC-DEM model (Jakobsen et al., 2000) strongly over-estimates both compressional
 285 and shear velocities because it consider the self-consistent scheme for the mixture of quartz and
 286 water (at the first procedure before replacing little by little water by hydrate). Such technique
 287 can be used for a highly compacted sandstone but it is not appropriate for the samples measured
 288 by Priest et al. (2009).

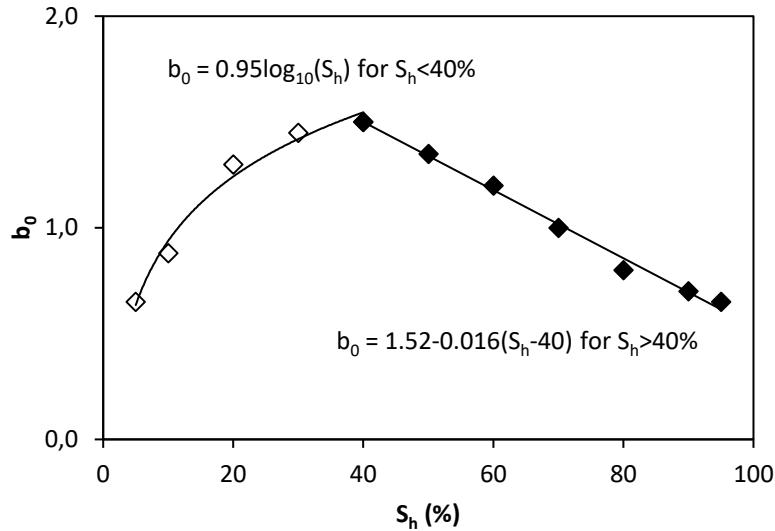


289
 290 *Figure 5: Validation of the model with experimental data of methane hydrate-bearing sand*
 291 *formed by the excess water method (Priest et al., 2009). Parameter: $c_0 = 0.1$.*

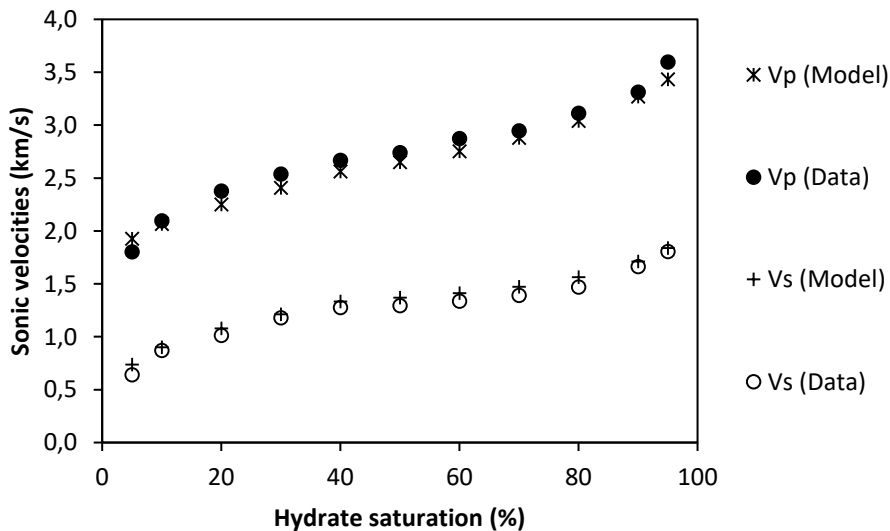
292
 293 The calibration of the relation (22) on the data of Priest et al. (2005) is limited to hydrate
 294 saturation lower than 40%. An additional calibration on data of Hu et al. (2012) shows that the
 295 relation between b_0 and S_h follows very well the trend defined by equation (22) for a hydrate
 296 saturation $S_h < 40\%$ but b_0 decreases linearly when S_h increases above 40% (*Figure 6*).
 297 Therefore, the following relation can be considered to predict b_0 for high degree of hydrate
 298 saturation:

$$b_0 = b_{40} - d_0(S_h - 40), \text{ for } S_h > 40\% \quad (24)$$

299 where b_{40} is the value of b_0 at $S_h = 40\%$ that is computed by equation (22) and d_0 a constant.
 300 It is shown on (Figure 7) that the sonic wave velocities computed using equations (17) to (24),
 301 with a calibrated parameter $c_0 = 0.95$ for $S_h < 40\%$ and a parameter $d_0 = 0.016$ for $S_h >$
 302 40% , fit very well the experimental data.
 303



304
 305 *Figure 6: Parameter b_0 versus hydrate saturation S_h : calibration using data measured by the*
 306 *test “run 18” of Hu et al. (2012).*



307
 308 *Figure 7 : A comparison between the proposed model and data measured by the test “run 18”*
 309 *of Hu et al. (2012). Parameters: $c_0 = 0.95$ and $d_0 = 0.016$.*
 310

311 **4.3. Applications for natural GHBS**

312 Konno et al. (2015) measured the compressional wave velocity of sediment samples
 313 containing methane hydrate cored under pressure from Japan offshore at about 300 ± 25
 314 meters below sea-bed. Measurement were performed in side a aluminium pressure housing
 315 (Suzuki et al. 2015). To apply the proposed model to these natural samples containing other
 316 minerals beside quartz contents, additional terms are added to the formulas (17) and (18) as
 317 follows

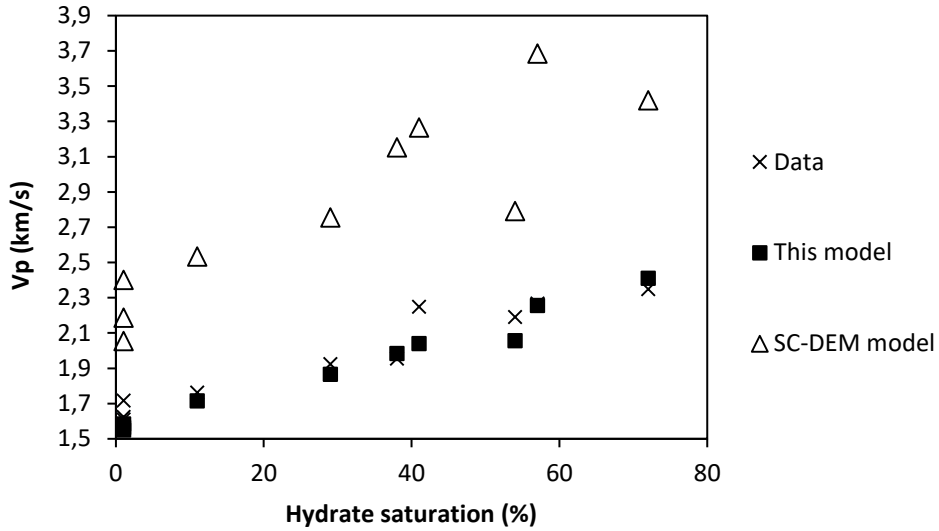
$$k_{hom} = \left[\frac{f_q}{k_q + a_0} + \frac{f_{other}}{k_{other} + a_0} + \frac{f_h}{k_h + a_0} + \frac{f_w}{k_w + a_0} \right]^{-1} - a_0 \quad (25)$$

318 and

$$\mu_{hom} = \left[\frac{f_q}{\mu_q + b_0} + \frac{f_{other}}{\mu_{other} + b_0} + \frac{f_h}{\mu_h + b_0} + \frac{f_w}{b_0} \right]^{-1} - b_0 \quad (26)$$

319 where the parameters $k_{other} = 21$ (GPa) and $\mu_{other} = 7$ (GPa) given by Konno et al. (2015)
 320 are considered. The weight fraction of quartz in the mineral phases as well as the densities of
 321 quartz and other mineral given by Konno et al. (2015) are used to compute the volume fraction
 322 of quartz f_q and other mineral f_{other} . *Figure 8* shows the prediction values versus experimental
 323 data of Konno et al. (2015). Note that the porosities of the measured samples range from 32 %
 324 to 47 %. Therefore each modeled point on *Figure 8* corresponds to one sample. The parameters
 325 $c_0 = 0.1$ and $d_0 = 0$ are used for the simulation to fit all the data. We compare also the present
 326 model with the existent SC-DEM model (Jakobsen et al., 2000; Hornby et al., 1994). One more
 327 time we observe that the existent SC-DEM model over-estimates the compressional sonic wave
 328 velocity of hydrate bearing sediment while the model presented herein fits very well with
 329 experimental data.

330

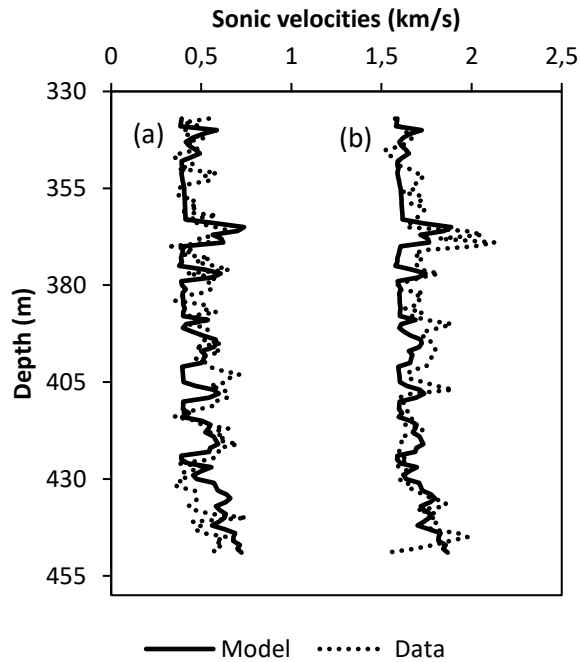


331

332 *Figure 8: Validation of the model with experimental data measured on natural methane*
 333 *hydrate-bearing shaly sand (Konno et al., 2015). Parameter: $c_0 = 0.1$ and $d_0 = 0$.*

334

335 *Figure 9 shows the validation of the proposed model on log data given by Chand et al. (2004)*
 336 *(where hydrate saturation was determined from electrical resistivity data and Archie's law). It*
 337 *can be observed that the present model can predict correctly the magnitude and the high-*
 338 *frequency variations of both V_p and V_s logs.*



339

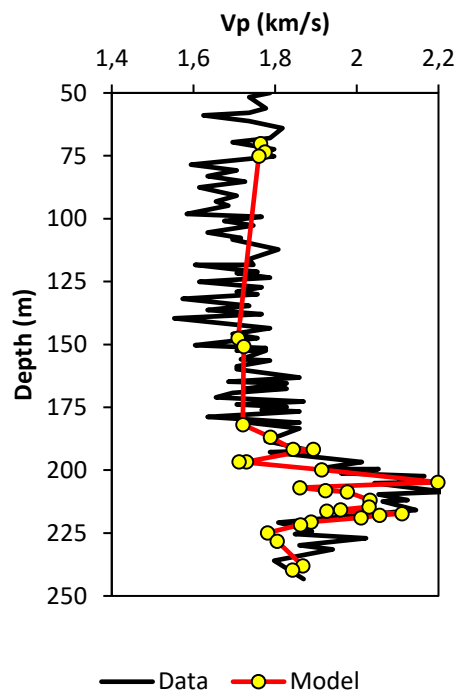
340 *Figure 9 : Validation of the model with log data of ODP Site 997, Blake Ridge, Leg 164 (see*
 341 *Chand et al., 2004, figure 12): (a) V_p and (b) V_s versus depth. Parameter $c_0 = 0.7$, the*

342 parameter d_0 is not needed because the hydrate saturation is lower than 40%. The points with
 343 S_h close to 1 (%), leading to a negative or diverse value of b_0 (computed using equation (22)),
 344 are not considered in the simulation.

345

346 Wang et al. (2014) used multi-channel seismic reflection data, well logs, and recovered
 347 sediments cores to characterize the geologic control on the occurrence of gas hydrate in the
 348 Shenhu area of the South China Sea. Data available from Wang et al. (2014) at one site (SH2)
 349 are used to predict the compressional velocity log in the present work. *Figure 10* presents the
 350 results obtained by the prediction (with $c_0 = 0.2$; $S_h < 40\%$ in this work) and those measured
 351 from the field. It can be seen that the prediction is in good agreement with the measured values.

352



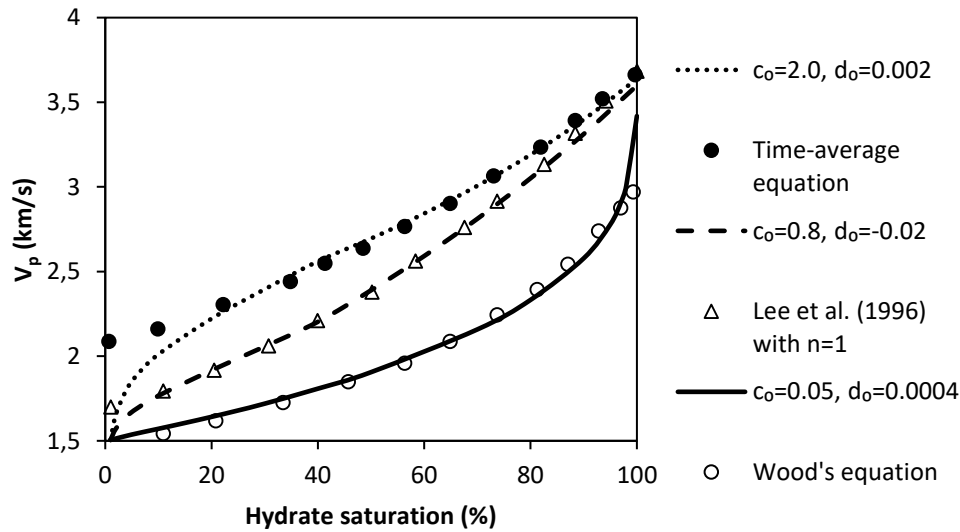
353

354 *Figure 10 : Validation of the model with log data in the South China Sea (Wang et al., 2014,*
 355 *Well SH2). Parameter $c_0 = 0.2$.*

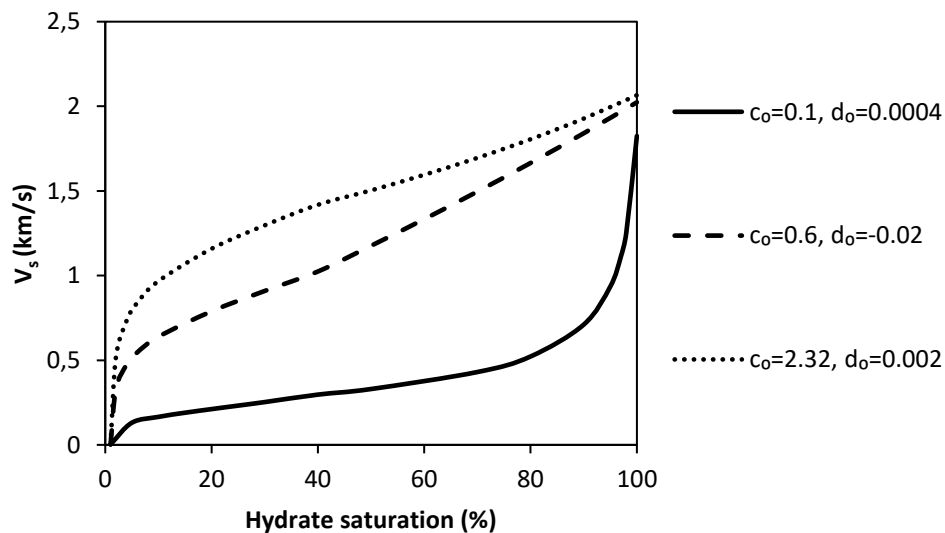
356

357 *Figure 11* shows a comparison between the proposed model and several existent key
 358 models given in literature: Wood's equation (Wood, 1941); time-average equation (Wyllie et
 359 al., 1958) as well as the model proposed by Lee et al. (1996). We observe that these existent
 360 models can be fitted by the present method just by changing the parameter c_0 . Consider for
 361 example a rock-bearing sandstone with an initial porosity of 57.5%, the Wood's method can be
 362 fitted by the proposed method herein by considering a parameter $c_0 = 0.05$ and $d_0 = 0.0004$.

363 Similarly, the time-average method can be fitted with $c_0 = 2.0$ and $d_0 = 0.002$. The results of
 364 Lee et al. (1996) with an exponent coefficient $n = 1$ can be fitted by using a parameter $c_0 =$
 365 0.8 and a negative parameter $d_0 = -0.02$ (i.e. b_0 increases when S_h increases above 40%). It
 366 is also verified that the results obtained by Lee et al. (1996) with a parameter $n > 1$ can be
 367 fitted with a parameter c_0 between 0.8 and 2.0 . It is of interest to remark that these existent
 368 models do not provide shear velocity while the present model provides both compressional and
 369 shear sonic velocities (*Figure 12*) with two calibration parameters c_0 and d_0 .



370
 371 *Figure 11 : Compression sonic velocity of GHBS versus its hydrate saturation with an initial*
 372 *porosity of 57.5%: a comparison between the proposed model and some existent models.*



373
 374 *Figure 12 : Shear sonic velocity of GHBS versus its hydrate saturation with an initial porosity*
 375 *of 57.5%.*

376

377 **4.4. Unsaturated GHBS**

378 Similar to the classical definitions of saturated or unsaturated rocks, here we employ the
 379 terms “partially saturated” or “unsaturated” for GHBS of which the pore space is not totally
 380 filled by water and hydrate but it is partially occupied by gas or air. Therefore the term
 381 “saturated” is used for GHBS with pore space totally filled by water and hydrate.

382 In section 4, we calibrate and validate a homogenization scheme that adapt with the
 383 microstructural change of saturated GHBS. With the case of unsaturated GHBS, it is necessary
 384 to add an additional terms related to air/gas phase in pore space to the formulas (17) and (18)
 385 as

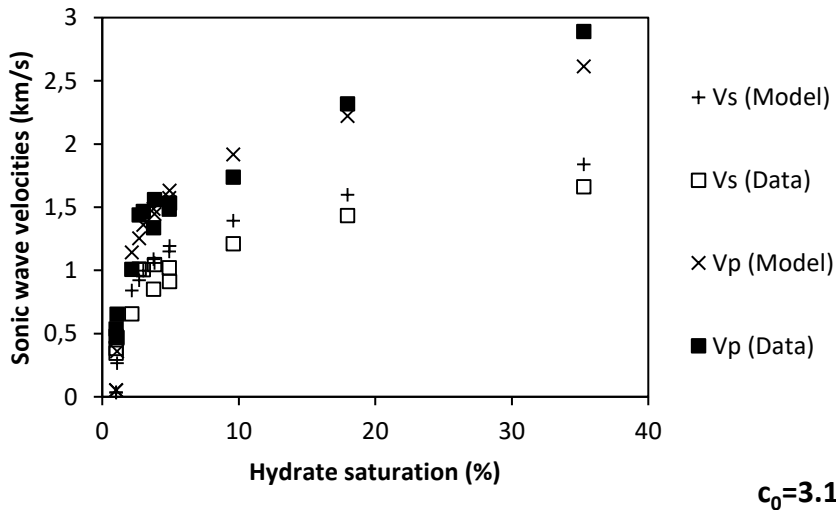
$$k_{hom} = \left[\frac{f_q}{k_q + a_0} + \frac{f_{gas}}{a_0} + \frac{f_h}{k_h + a_0} + \frac{f_w}{k_w + a_0} \right]^{-1} - a_0 \quad (27)$$

$$\mu_{hom} = \left[\frac{f_q}{\mu_q + b_0} + \frac{f_{gas}}{b_0} + \frac{f_h}{\mu_h + b_0} + \frac{f_w}{b_0} \right]^{-1} - b_0 \quad (28)$$

386 The volume fraction of the phases are calculated by

$$f_q = 1 - \phi; f_{gas} = \frac{S_{gas}}{100} \phi; f_h = \frac{S_h}{100} \phi; f_w = 1 - f_q - f_h - f_{gas} \quad (29)$$

387 Note that equation (20) for the calculation of the bulk density remains valid due to almost zero
 388 density of air/gas phase. *Figure 13* shows a good fit between the prediction and data of dry gas
 389 hydrate sand measured by Priest et al. (2005) (direct measured data on dry sample, i.e. before
 390 transforming thought Gassmann calculation).



391
 392 *Figure 13:* Sonic velocity of dry gas hydrate-bearing sand: Experimental data (Priest et al.
 393 2005) versus prediction by the present model.

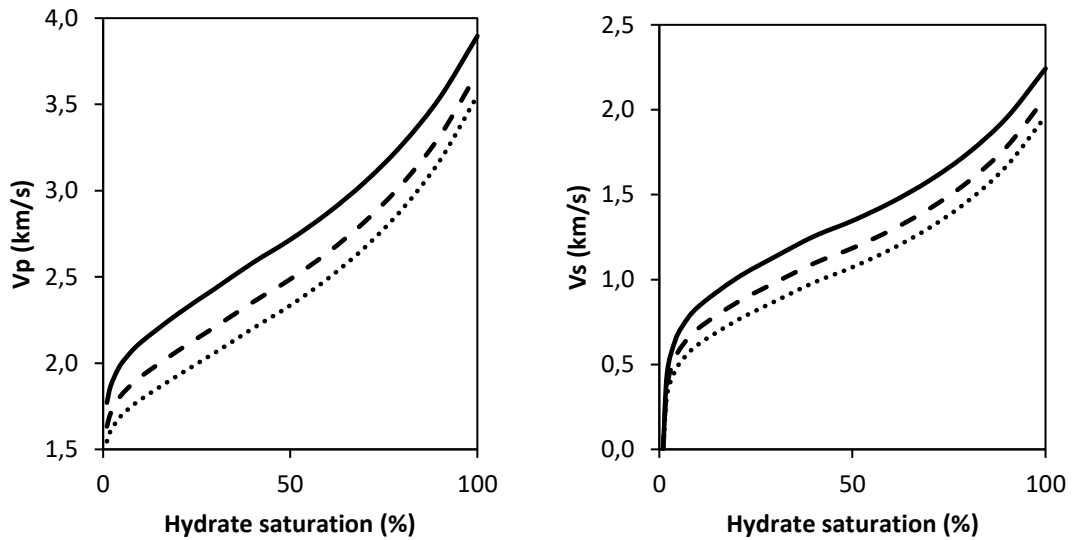
394

395 **4.5. Sensitivity analysis**

396 **4.5.1. Influence of the initial porosity**

397 *Figure 14* shows the sensitivity of the initial porosity to the relationship between
398 compression and shear sonic velocities versus hydrate saturation with fixed parameters $c_0 =$
399 0.6 , $d_0 = 0$. For a porosity varies from 30% to 50% the compression sonic velocity decreases
400 of about 10% to 15% and the shear velocity decreases of about 15% to 30% those are quite
401 significant. However *Figure 14* shows that the sensitivity of the porosity to the relationship
402 between the compression and the shear velocities are negligible, especially for a shear velocity
403 higher than 1 (km/s) that corresponds to a hydrate saturation higher than 20%. The later explains
404 a quite narrow trend of field data shown on *Figure 2* (filled circle points).

405

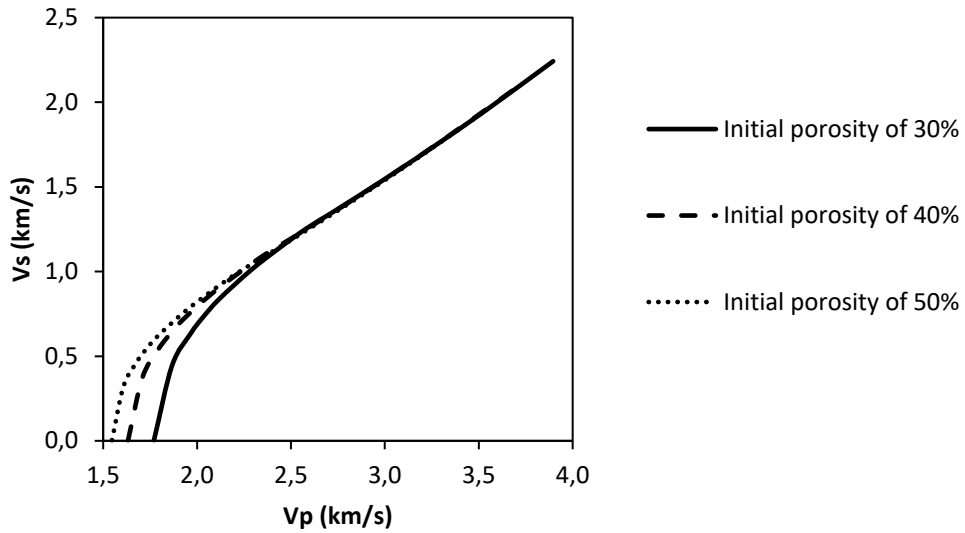


406

407

(a)

408



409

410

(b)

411 *Figure 14 : Influence of the initial porosity on (a) Sonic velocities of GHBS versus its hydrate*
 412 *saturation; (b) Relationship between shear and compressional sonic velocities of GHBS.*

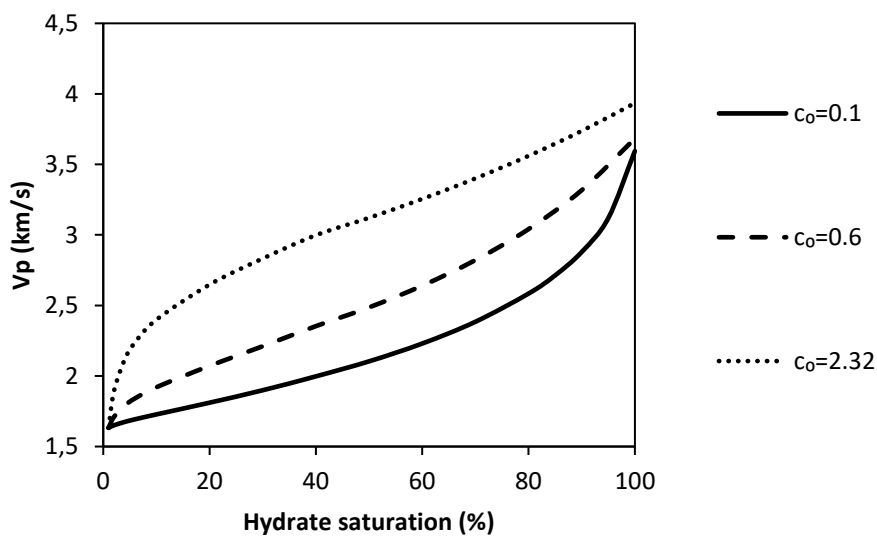
413

414 4.5.2. Influence of the parameter c_0

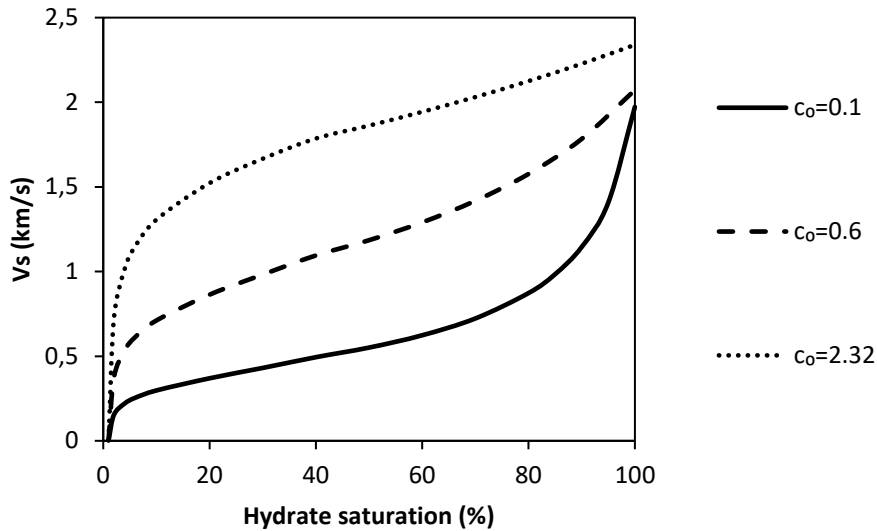
415 *Figure 15* shows a strong effect of the parameter c_0 on the relationships between the
 416 compressional and shear sonic velocities versus hydrate saturation with a porosity of $\phi = 0.4$
 417 and a fixed parameter $d_0 = 0$. Therefore, to predict the hydrate saturation from sonic logs, it
 418 is important to correctly calibrate the parameter c_0 . *Figure 16* shows that the relation V_p-V_s is
 419 also dependent on the parameter c_0 .

420

421

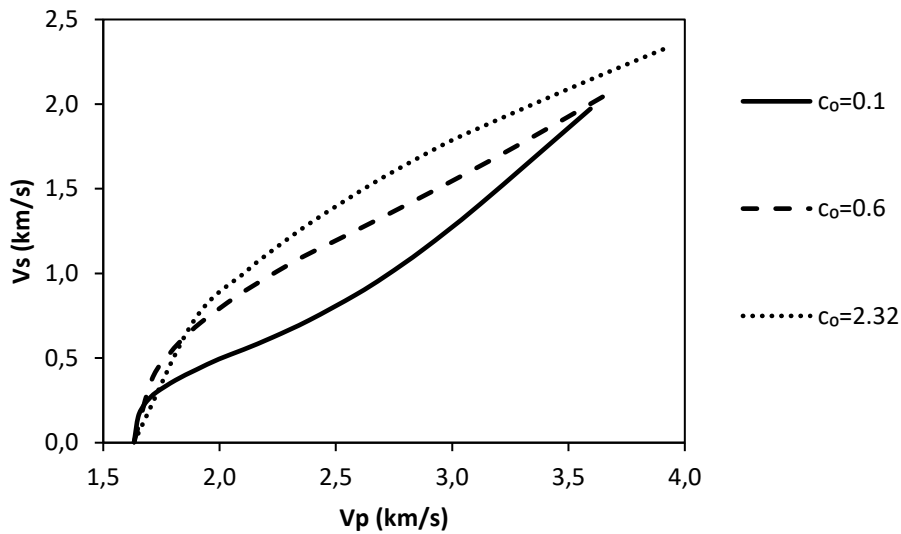


422



423
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 428

Figure 15 : Sonic velocities of GHBS versus its hydrate saturation for different values of c_0 in the range between 0.10 and 2.32 those are previously calibrated from laboratory data. Fixed parameters: $\phi = 0.4$ and $d_0 = 0$.



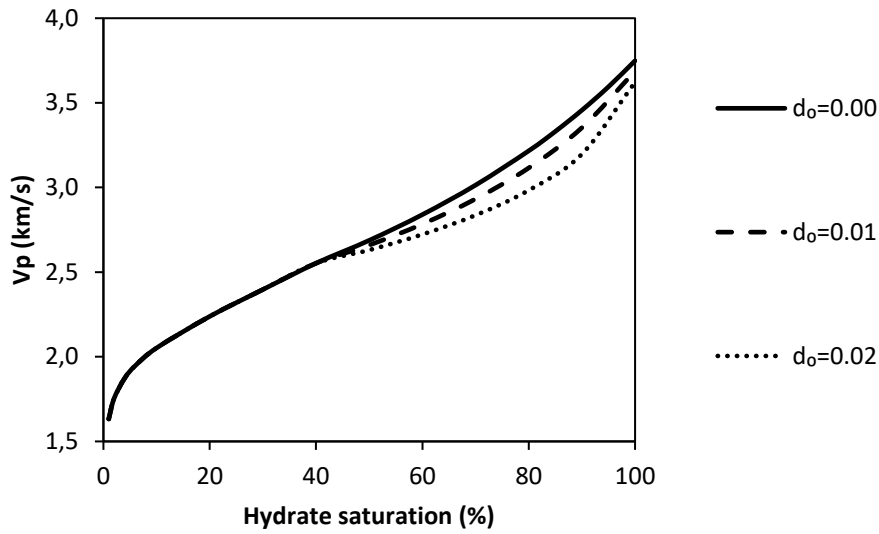
429
 430
 431
 432

Figure 16 : Influence of the parameter c_0 on the relationship between shear and compressional sonic velocities of GHBS. Fixed parameters: $\phi = 0.4$ and $d_0 = 0$.

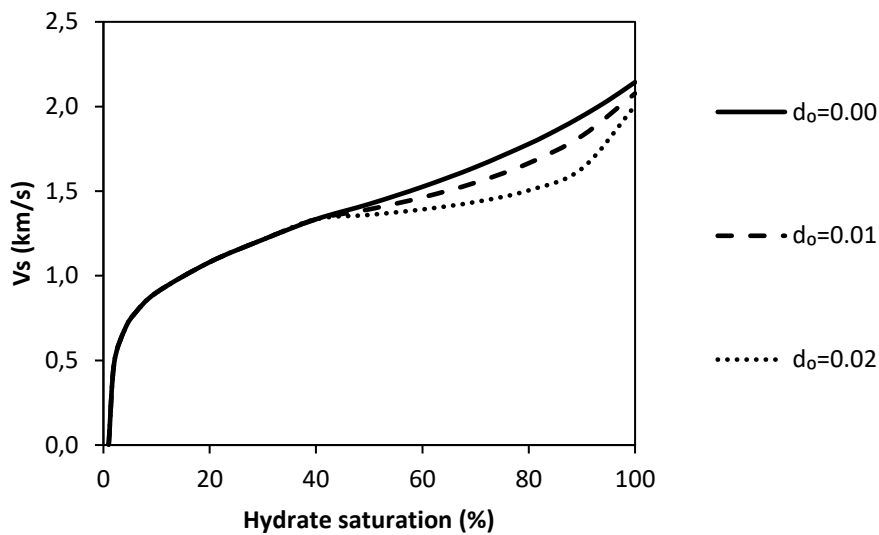
433 4.5.3. Influence of the parameter d_0

434 Figure 17 and Figure 18 show a quite weak effect of the parameter d_0 on the
 435 relationships between the compressional and shear sonic velocities versus hydrate saturation
 436 with fixed parameters $\phi = 0.4$ and $c_0 = 1.0$. Besides, the in-situ hydrate saturation is usually

437 in the range below 40%. Therefore, the role of the parameter d_0 is less important than the
438 parameter c_0 .



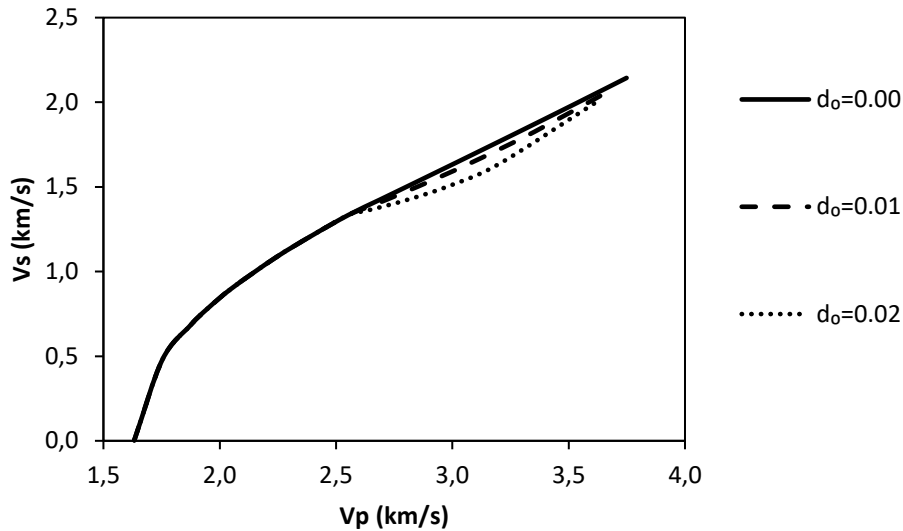
439



440

441 *Figure 17 : Sonic velocities of GHBS versus its hydrate saturation for different values of d_0 in*
442 *the range between 0 and 0.02 those are previously calibrated from data. Fixed parameters:*
443 *$\phi = 0.4$ and $c_0 = 1.0$.*

444



445
 446 *Figure 18 : Influence of the parameter d_0 on the relationship between shear and compressional*
 447 *sonic velocities of GHBS. Fixed parameters: $\phi = 0.4$ and $c_0 = 1.0$.*

448 5. Concluding remarks

449 An adaptive homogenization scheme of which the elastic properties of the reference matrix
 450 are linearly related to the logarithm of hydrate saturation is proposed and calibrated for gas
 451 hydrate-bearing sand. The developed model can predict measured data just by changing two
 452 parameters c_0 and d_0 . The parameter c_0 strongly depends on the morphology of the gas-hydrate
 453 phase. More precisely, c_0 is in a high range (about 2.0 to 3.0) if the gas-hydrate phase plays the
 454 role of the cement-phase that increases the stiffness of the rock frame. Inversely, for the case of
 455 weak GHBS, c_0 is in a low range (around 0.1) if the gas-hydrate phase plays the role of a pore-
 456 filling phase. Natural GHBS corresponds to a value in between these limits. Indeed, Nguyen et
 457 al. (2015) showed that an uncemented sand packing with a given porosity ϕ is as weak as a
 458 cemented one with a porosity of about 2ϕ .

459 The proposed model is validated with various experimental data obtained in laboratory
 460 including gas hydrate-bearing sands formed in laboratory, natural GHBS containing multi
 461 minerals. In addition, the prediction is also compared with the log data taken from various sites.
 462 All these validations evidence the robustness of the model.

463 When comparing the proposed model to the existing models, the proposed model is quite
 464 simple with two calibration parameters c_0 and d_0 . In addition, it can predict both compression
 465 and shear sonic velocities within these parameters.

466 The results in the present work suggest that the calibration parameter c_0 would reflect the
 467 morphology of the GSBS. It is in a high range (around 2) if gas hydrates play the role of cement

468 phase while it is in a low range (around 0.1) if gas hydrates locate at the pore space between
 469 solid particles. The present model offers an easy and accurate technique to predict the hydrate
 470 saturation in GHBS using sonic logs and/or seismic acquisition.

471

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 474 Agency for funding the present study, which is part of the project HYDRE “Mechanical
 475 behavior of gas-hydrate-bearing sediments” –ANR-15-CE06-0008. Related data of the paper
 476 are accessible by using the DOI: [10.6084/m9.figshare.6167717](https://doi.org/10.6084/m9.figshare.6167717).

477

478 **Appendix: the combined self-consistent and DEM method (Jakobsen et al., 2000; Hornby 479 et al., 1994)**

480 For low hydrate saturation, water is first mixed with the solid phase via the self-
 481 consistent scheme (see also Eqs. (15) to (18))

$$k_{SC} = \left(\frac{\phi}{k_w + a_{SC}} + \frac{1 - \phi}{k_q + a_{SC}} \right)^{-1} - a_{SC} \quad (30)$$

482 and

$$\mu_{SC} = \left(\frac{\phi}{\mu_w + b_{SC}} + \frac{1 - \phi}{\mu_q + b_{SC}} \right)^{-1} - b_{SC} \quad (31)$$

483 with

$$a_{SC} = \frac{4}{3}\mu_{SC}; \quad b_{SC} = \frac{\mu_{SC}(9k_{SC} + 8\mu_{SC})}{6(k_{SC} + 2\mu_{SC})} \quad (32)$$

484 Once k_{SC} and μ_{SC} are determined, hydrate considered via a multi-step differential effective
 485 medium technique (DEM) that allows replacing little-by-little water in the pore space by
 486 hydrate. At a step i , the bulk and shear moduli can be computed by

$$k_{i+1} = \left[\frac{1}{k_i + a_i} + \frac{dv}{k_h + a_i} - \frac{dv}{k_w + a_i} \right]^{-1} - a_i \quad (33)$$

487 and

$$\mu_{i+1} = \left[\frac{1}{\mu_i + b_i} + \frac{dv}{\mu_h + b_i} - \frac{dv}{b_i} \right]^{-1} - b_i \quad (34)$$

488 where dv the increment of volume of water that is replaced by hydrate. The parameters a_i and
 489 b_i have a similar form of Eq. (9) as

$$a_i = \frac{4}{3}\mu_i; b_i = \frac{\mu_i(9k_i + 8\mu_i)}{6(k_i + 2\mu_i)} \quad (35)$$

490 At the first step, the effective bulk and shear moduli obtained by Eqs. (30) to (32) are used such
 491 as $k_0 = k_{SC}$ and $\mu_0 = \mu_{SC}$.

492 For high hydrate saturation (>50%), the effective bulk and shear moduli are calculated
 493 by a similar ways as the case of low hydrate saturation but the role of water and hydrate are
 494 interchanged. More precisely, the self-consistent procedure is first employed to mixe hydrate
 495 with the solid phase assuming 100% hydrate saturation. Then the DEM procedure is used to
 496 replacing litle by litle hydrate by water until reaching the desired hydrate saturation.

497

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