A bond contact model for methane hydrate bearing sediments with inter-particle cementation

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Abstract

While methane hydrate (MH) can be present in various forms sands deep seabeds or permafrost regions, this paper deals with methane hydrate bearing sediments (MHBS) where the MH has formed bonds between sand grains. A bond model based on experimentally-validated contact laws for cemented granules is introduced to describe the mechanical behavior of the MH bonds. The model parameters were derived from measured values of temperature, water pressure and MH density. Bond width and thickness adopted for each bond of the MHBS were selected based on the degree of MH saturation. The model was implemented into a 2D-DEM code. A series of numerical biaxial standard compression tests were carried out for various degrees of MH saturation. Comparison with available experimental data shows that the model can effectively capture the essential features of the mechanical behavior of MHBS for a wide range of levels of hydrate saturation. In addition, the analyses here presented shed light on the relationship between level of cementation and de-bonding mechanisms taking place at the microscopic level and the observed macro-mechanical behavior of MHBS. Also the analyses shed light on the relationship between spatial distribution of bond breakages and contact force chains with the observed strength, dilatancy and deformability of the samples.

Keywords: methane hydrate; cementation; bond contact model; discrete element method; biaxial compression tests

1 Introduction

Methane hydrate (MH) is regarded as one of the most promising resources to alleviate current and future energy needs [1-4]. MHs usually form in deep seabeds and permafrost regions where MHs can remain stable under low temperatures and high pressures [5-8]. They can greatly enhance the strength of the host sediments [9]. However, changes in pressure and temperature conditions and human interventions (i.e., installation of offshore pipelines, cables and platforms) may cause MH dissociation with the consequent destabilization of methane hydrate bearing sediments (MHBS) and lead to large marine landslides. Unfortunately, the mechanisms originating these geo-hazards are still poorly understood due to a lack of knowledge of the fundamental mechanical properties of MHBS. This also makes it difficult to establish a universal constitutive model for MHBS [8], although some models were proposed in the literature showing capability in capturing several basic features of MHBS (e.g., [10, 11]), and were employed to solve boundary value problems using either the finite element method (FEM) or the finite difference method (FDM) (e.g., [12]).

The formation of MHs is affected by a few factors such as host deposit, gas percolation speed and path, and initial water saturation [13, 14], leading to a variety of micro structures in MHBS. As illustrated in Figure 1, Waite et al. [15] identified three main formation habits at the pore scale: (1) pore-filling, with MHs floating in the pore fluid without bridging any particles; (2) load-bearing, with hydrate particles taking part in the strong force chains of the granular assembly; and (3) cementation, with

MHs cementing sand grains (acting as bond bridges between grains). As suggested by Waite et al. [16] and confirmed by experimental data from different sources(e.g., [13, 17, 18]), the mechanical properties of MHBS strongly depend on the formation habits of the hydrates. For instance, hydrates acting as cementation agents at inter-particle contacts give rise to larger values of strength and stiffness for the MHBS than pore-filling hydrates. This implies that hydrate morphology has to be taken into account in any realistic constitutive models of MHBS. Only an investigation at the pore scale can establish the link between pore habits and macroscopic properties of MHBS (e.g., [19]).

The distinct element method (DEM), originally proposed by Cundall and Strack [20] for dry granules, has significant potential in shedding light on the relation between pore habit of the hydrates and bulk properties of MHBS by modeling hydrate morphology at the grain scale. Recently, DEM has been employed to investigate some significant features of MHs, such as hydrate growth [21], hydrate dissociation [22], hydrate distribution of pore-filling patterns [8, 23, 24] or of cementation patterns [25, 26]. Brugada et al. [8] investigated the mechanical properties of MHBS with MH in the form of pore-filling habit via three-dimensional DEM simulations. Discrete spheres one order of magnitude smaller than the soil particles were randomly generated and distributed within the voids to replicate hydrate particles. In their simulations, soil grains and MH particles get into contact and exchange forces, however without exhibiting any cementation at contacts. In that work, the contact stiffnesses of the hydrate particles were determined on the basis of a parametric study.

Jung et al. [23, 24] characterized mechanical behaviors of MHBS in two cases: distributed hydrates and patchy saturation. In these works hydrate particles were randomly generated in space and attached to the sand grains so that their effect on the load bearing force chains could be investigated. However, few researches have been carried out on the cementation pattern of MHs due in part to a lack of properly validated bond contact models for MHBS accounting for essential factors such as hydrate saturation, temperature, and water pressure. Although some early attempts were made to model MHs in the cementation habit via DEM, the previous models were over-simplified without clarifying the exact correlation between hydrate saturation and bond strength [25], or neglecting the role played by the thickness of the MH bonds [26]. However, MHs forming bonds between grains of the host granular soil are of common occurrence since MHs are prone to cement unconsolidated sediments containing an abundant gas phase. For example, in the Blake Ridge off the southeast coast of the United States [27] and in the Cascadia margin [28], MHs have been recognized in formation habit as cementing. In this case, the formation of MHs causes the onset of hydrate bonds between nearby sand grains at the pore scale. Experimental data [14, 16, 29-32] show that this pore habit plays an important role in the macro-mechanical behavior of MHBS, and in particular substantially affects the bulk properties of MHBS far more than the pore-filling habit. Therefore there is a need to investigate the influence of MH bonds on the bulk properties of the host sediments. To achieve this goal via DEM simulations, a suitable bond contact model accounting for all the significant factors affecting the bond behavior is of critical importance. Establishing a suitable bond model also paves the way to use DEM analyses to investigate the effect of time dependent phenomena taking place within inter-particle bonds on MHBS. In fact, chemical reactions may occur over time with the effect of strengthening or weakening the bonds (aging effects). The DEM could be employed to investigate these time-dependent phenomena.

It has been widely recognized that inter-particle bonds in some other cemented materials, such as sandstone, mortars, grouted soils and volcanic ashes, affect many aspects of soil behavior, e.g., enhancing strength and shear dilation [33, 34]; ruling strain softening [35-41]; and influencing the formation of shear bands [38, 42]. The inter-particle bonds of these cemented soils are mainly made of Portland cement, gypsum and lime owning properties different from those of MHs. Little is known about MH bonds which, however, largely affect the macro-mechanical behavior of MHBS. Hence, it is authors' opinion that research on the macro-mechanical properties of MHBS should be informed by its microstructure and the behavior observed at the micro scale. Scanning electron microscopy (SEM) images and X-ray CT images have been used to obtain morphological information of MHBS at the scale of the grain. However, these techniques alone are insufficient to gain a comprehensive understanding of the macro mechanical response of MHBS, particularly in relation to the bonding effects of MHs.

This paper aims to establish a suitable bond contact model for sandy deposits with MH bonds based on an experimentally-validated contact law achieved for cemented granules. Following this introduction, a general bond contact model derived from a series of micro experiments is introduced with particular emphasis on the bond strength envelope. Section 3 extends the generic model to MHBS by relating the model parameters to the surrounding temperature, water pressure, and MH density in addition to the hydrate saturation. DEM implementation of this model is described in Section 4 followed by relevant simulation results to be presented in Section 5 in comparison to the available experimental data [18].

2. A bond contact model for cemented granules

Figure 2 illustrates conceptually the 2D bond contact model adopted in the DEM simulations presented in this paper. The bond between two disks of radii R_1 and R_2 has a finite width, B, and a finite thickness varying along the disk surface. The bond thickness is hereafter characterized by its measure at the center of the bond, t_0 (see Figures 2(a)). Figures 2(b) to (d) provide the mechanical responses of this model in three directions (i.e., normal, tangential and rolling directions). As illustrated in the figures, the force-displacement and the moment-rotation laws are featured by an initially linear elastic response, brittle breakage and perfect plastic behavior. This laws have been experimentally derived from bonds made of either epoxy resin [43] or cement [44].

For an intact bond, the normal force F_n, the shear force F_s, and the moment M can be computed as:

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

$$F_s = K_s u_s, \tag{1b}$$

$$M = K_r\theta, \tag{1c}$$

where u_n, u_s, and θ are the overlap, relative shear displacement, and relative rotation angle, respectively, whilst K_n, K_s, K_r are the normal, tangential and rolling bond contact stiffness, respectively. u_0 is the distance between two adjacent particles at the time of formation of the bond. In the case that two particles are in contact during the formation of the bond, the minimum bond thickness, t_0, is assumed to be 0 and $u_0 = 0$. To account for the distance existing between particles at the time of formation of the MH bonds is important for a realistic modeling of MHBS since this has an influence on the observed mechanical behavior of the assembly of bonded particles.

The thresholds (or bond strengths) in the normal, shear and rolling directions are here denoted by R_{nb}, R_{sb} and R_{rb}, which define the upper bound of the elastic regime in each direction. The bond will break in a fragile fashion if any force or moment exceeds these thresholds. When two grains after their bond has broken come into contact again, the contact laws between un-bonded grains are as follows:

$$F_n = K_n^{'} u_n, \tag{2a}$$

$$F_s = \min\left[K_s^{'} u_s,\ \mu F_n\right], \tag{2b}$$

$$M = \min\left[K_r^{'}\theta,\ \frac{F_n \cdot \delta \cdot \bar{R}}{6}\right], \tag{2c}$$

where min[·] is the operator taking the minimum value; μ is the inter-particle friction coefficient; $\bar{R} = 2R_1R_2/(R_1+R_2)$ is the equivalent radius of two particles in contact. $K_n^{'}$, $K_s^{'}$ and $K_r^{'} = K_n^{'}(\delta\bar{R})^2/12$ are the normal, tangential and rolling contact stiffnesses between soil particles in contact which are different from the stiffnesses introduced in Eqs. (1) which account for both bond and particle stiffnesses. δ is the shape parameter of soil particles characterizing the rolling resistance of particles in contact, which is meant to account for the effect of non-spherical grain shapes. Details on the physical meaning of the inter-particle rolling resistance can be found in Jiang et al. [45].

In case of combined loads, the adopted strength criterion is represented as a three-dimensional surface in the F_n-F_s-M space. According to experimental results on epoxy resin and cement [43, 44], the slice of the surface in the F_s-M plane can be described in first approximation by an ellipse:

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

where R_{sb0} is the bond shear strength in the absence of bending moments and R_{rb0} is the bond rolling strength in the absence of shear forces. The size of the ellipse depends on F_n. Hence both R_{sb0} and R_{rb0} depend on the magnitude of F_n:

$$R_{sb0}=f_s\cdot L_s\cdot(F_n+R_{tb})\cdot\left(\ln\frac{R_{cb}+R_{tb}}{F_n+R_{tb}}\right)^n, \tag{4a}$$

$$R_{rb0}=f_r\cdot L_r\cdot(F_n+R_{tb})\cdot\left(\ln\frac{R_{cb}+R_{tb}}{F_n+R_{tb}}\right)^m, \tag{4b}$$

where R_{tb}, R_{cb} are the bond tensile and compressive strength, respectively, which can be obtained from pure tension and compression tests on the cemented granules. L_s, L_r are the slope of the straight lines linking R_{tb} to the peak shear strength or rolling resistance on the projection plane (see Figure 3). Coefficients f_s, f_r, n and m are fitting parameters calibrated on the available experimental data. A comparison between the curves of Equation (4) and available experimental data, [43, 44], is shown in Figure 3. A good agreement between the curves and the experimental data is apparent. Combining Equations (3) and (4) gives rise to a three dimensional strength envelope shown in Figure 4 which has proved to replicate satisfactorily the available experimental data for combined loading paths too.

The contact law for an intact bond requires knowledge of the bond strength and stiffness parameters summarized in Table 1. For cement- or epoxy-bonded case, these parameters can be directly obtained from microscopic mechanical tests [43, 44]. For the MHBS case, however, it is very difficult to conduct micro-mechanical tests on MH bonded granules because of the extreme experimental conditions required, e.g., very low temperature and very high pressure, which make it very difficult to obtain reliable direct measurements of the parameters of the MH bonds. Jung and Santamarina [46] performed experimental tests at the micro scale to measure adhesive and tensile strengths of two MH-bonded flat smooth surfaces of mica and calcite; however their measurements are not directly applicable to the bonds considered here since the bond strength is likely to be significantly affected by the curvature of the surfaces of grains near their contact point, grain surface roughness and impurities in the MH. Thus, it is necessary to introduce some assumptions to determine indirectly the model parameters subsequently illustrated here.

3. Extension of the bond contact model to MHBS

Concerning MHBS located at a depth of *h* below sea level, Figure 5(a) provides a sketch of two MH bonded granules at the temperature *T*, and the surrounding water pressure σ_w. In this section, the model parameters (i.e. bond strength and bond contact stiffness) relevant to MH bonds will be indirectly determined with respect to the surrounding environment. Moreover, the geometric features (e.g, the width and thickness) of the MH bonds will be studied considering the MH saturation and MH formation characteristics in the host sands.

3.1 Bond contact stiffness of MH

As shown in Equation (1), the bond contact stiffness can be characterized by K_n, K_s, K_r in the normal, shear and rolling direction, respectively. In general, the Young's modulus for soil grains ranges from 50 to 70 GPa. They can be regarded as rigid particles when the bonds (e.g., MH bonds) have relatively lower elastic modulus. As shown in Figure 2, for the case of rigid particles with deformable elastic bond, the normal deformation of the bond material, δ_n, can be expressed as:

$$\delta_n = \varepsilon t = \frac{\sigma t}{E} = \frac{F_n t}{BE}, \tag{5}$$

where ε is the normal strain; σ is the evenly distributed normal stress; E is the Young's modulus of the bonding material and t is the average thickness of the bonding material (see Figure 2). Thus the normal contact stiffness can be related to E as follows:

$$K_n = \frac{F_n}{\delta_n} = \frac{BE}{t}. \tag{6}$$

According to experimental data obtained from tests on pure MHs (e.g., [47]), E is strongly related to the temperature, T, the confining pressure, (i.e., pore pressure in this case, σ_w), and MH density, ρ. The relationship can be written as:

$$E = E(\sigma_w, T, \rho) \quad . \tag{7}$$

Figures 6(a) and (b) present the collected stress-strain response curves obtained from triaxial compression tests on pure MH samples under different testing temperatures and confining pressures [47]. The information available on MH density is also presented for each curve. The Young’s modulus of MH under different testing conditions, i.e., the tangential modulus at the point where the deviator stress is one half of the peak value, was obtained and listed in Table 2, which was used to produce Figure 7 in order to show the factors influencing the Young’s modulus of MH. As shown in Figs. 7(a) and (b), the Young’s modulus increases linearly with the MH density at a rate which is not significantly affected by confining pressure and temperature. Data in Figs. 7(a) and (b) can be regrouped for different MH densities (i.e., 0.7, 0.8 and 0.9 g/cm^3) resulting in Figs. 7(c) and (d) that illustrate the variation of the Young’s modulus of MH with respect to confining pressure and temperature. Figures 7(c) and (d) indicate that the Young’s modulus of MH linearly increases with confining pressure and linearly decreases with temperature at a rate which is significantly affected by the MH density. Hence we assume absence of coupling between the three factors: confining pressure, temperature and MH density. The Young's modulus of MH can be expressed as:

$$\frac{E}{p_{\mathrm{a}}} = a_0 + a_1\left(\frac{\sigma_w}{p_{\mathrm{a}}}\right) + a_2\left(\frac{T}{T_0}\right) + a_3\left(\frac{\rho}{\rho_w}\right), \tag{8}$$

where p_a is the standard atmospheric pressure (i.e., 1.01×10^5 Pa); T_0 is a reference temperature of 1 ˚C; ρ_w is the density of water at the temperature of 4˚C; a_0, a_1, a_2, and a_3 are constant coefficients achieved by fitting the data in Table 2. As a result, Equation (8) yields:

$$\frac{E}{p_{\mathrm{a}}} = 3\left(\frac{\sigma_w}{p_{\mathrm{a}}}\right) - 1.98\left(\frac{T}{T_0}\right) + 4950.50\left(\frac{\rho}{\rho_w}\right) - 1821.78. \tag{9}$$

The values of Young modulus obtained by using Equation (9) are listed in Table 2 for sake of comparison with the available experimental data. A good agreement is apparent. The tangential and rolling bond contact stiffnesses can be assumed to be proportional to the normal contact stiffness, with the former one determined by experimental investigations [43, 44] and the latter one determined by the assumption of elastic bond, [36]:

$$K_s = \frac{2}{3} K_n \ . \tag{10}$$

$$K_m = \frac{1}{12} K_n B^2. \tag{11}$$

3.2 Bond strengths of MH

3.2.1 Bond tensile/compressive strength

When a tension or compression force is applied, the normal force is assumed to be evenly distributed over the whole width of the inter-particle MH. Hence, tensile and compressive strengths of MH bonds, i.e. R_{tb} and R_{cb} in Equations (4), can be computed from the tensile and compressive strengths of a pure MH specimen, $q_{\max,t}$ and $q_{\max,c}$ respectively, subject to a given confining pressure σ_w:

$$R_{tb} = B \times q_{\max,t}, \tag{12a}$$

$$R_{cb} = B \times q_{\max,c}, \tag{12b}$$

Figure 5(b) provides the shear strength envelope of pure MH having assumed the validity of the Mohr-Coulomb failure criterion. Given a prescribed pore water pressure, σ_w, the minimum and maximum principal stresses are σ_t and σ_c, respectively. Depending on the value of water pressure, σ_t may also be tensile (i.e. negative according to the soil mechanics stress convention). Thus, $q_{\max,t}$ and $q_{\max,c}$ can be expressed as:

$$q_{\max,t} = (\sigma_w - \sigma_t), \tag{13a}$$

$$q_{\max,c} = (\sigma_c - \sigma_w), \tag{13b}$$

The maximum deviator stress $q_{\max,c}$ obtained from a compression triaxial test on a pure MH specimen (without any soil grains included) strongly depends on the temperature, T, confining pressure , σ_w, and MH density , ρ [47]. Hence it can be expressed as:

$$q_{\max,c} = q_{\max,c}(\sigma_w, T, \rho), \tag{14}$$

so that in the light of Equations (12), $R_{tb} = R_{tb}(\sigma_w, T, \rho)$ and $R_{cb} = R_{cb}(\sigma_w, T, \rho)$. Figures 8(a), (b) and (c) illustrate the variation of the maximum deviator stress against pore water pressure, temperature and MH density based on published experimental data [47]. In Figure 8 (b), the solid line and dash line represent the experimental results of MH with high and low purity, respectively. Results of MH at high purity were selected in our analysis. Since the tests on MH with high purity were only performed at T=-30 °C, the dash line is plotted assuming that it exhibits the same tendency as that of the solid line. As shown in these figures, the maximum deviator stress of MH linearly increases with the MH density and the confining pressure, and linearly decreases with the temperature. For sake of simplicity, we neglected the reciprocal influence of the three factors on the maximum deviator stress of MH. Thus, Equation (14) can be expressed by:

$$\frac{q_{\max,c}}{p_{\mathrm{a}}}=b_0+b_1\left(\frac{\sigma_w}{p_{\mathrm{a}}}\right)+b_2\left(\frac{T}{T_0}\right)+b_3\left(\frac{\rho}{\rho_w}\right), \tag{15}$$

where b_0, b_1, b_2, and b_3 are fitting parameters. Table 3 lists the available experimental data of MH with high purity [47] and the data attained from the dash line in Figure 8(b). Fitting the data in Table 3, Equation (15) yields:

$$\frac{q_{\max,c}}{p_{\mathrm{a}}}=0.81\left(\frac{\sigma_w}{p_{\mathrm{a}}}\right)-2.08\left(\frac{T}{T_0}\right)+184.16\left(\frac{\rho}{\rho_w}\right)-134.65. \tag{16}$$

The prediction based on Equation (16) is also given in Table 3 for sake of comparison with the available experimental data. A good agreement is apparent. Equation (16) is assumed to also hold for the extension triaxial test, leading to the following:

$$\frac{q_{\max,t}}{p_{\mathrm{a}}}=0.81\left(\frac{\sigma_t}{p_{\mathrm{a}}}\right)-2.08\left(\frac{T}{T_0}\right)+184.16\left(\frac{\rho}{\rho_w}\right)-134.65. \tag{17}$$

Combining Equation (13a) and Equation (16) to cancel out σ_t, Equation (17) can be re-written as:

$$\frac{q_{\max,t}}{p_{\mathrm{a}}}=0.45\left(\frac{\sigma_w}{p_{\mathrm{a}}}\right)-1.15\left(\frac{T}{T_0}\right)+101.75\left(\frac{\rho}{\rho_w}\right)-74.39. \tag{18}$$

3.2.2 Shape of the strength envelope

As demonstrated in Figure 3, the shape of the envelope is controlled by the cementation materials, resulting in different values of fitting parameters in Equation (4). Direct calibration of these parameters from laboratory tests for MH bonds in the same manner as cement or epoxy bonds [43, 44] is extremely difficult. So at present the mechanical properties of MH-bonded sand grains can only be inferred by standard macroscopic geotechnical tests and analogy to similar types of materials. In Figure 9, the yielding curves of different materials in the normalized $(\sigma_1 - \sigma_3)$ and σ_3 plane are presented. The yielding curve is 'right skewed' with a right tail for cement-based material and 'left skewed' for epoxy resin. Unfortunately, the yielding curve of MH cannot be inferred from the scanty experimental data available. However, MH is an ice-like material composed of methane gas and water, i.e., natural gas is trapped inside cage-like crystal structures made up of water molecules [7]. Its physical [48, 49] and mechanical [50, 51] properties have been found to be similar to those of ice. Therefore the yielding curve of ice has been plotted in Figure 9(b) for comparison. The curve relative to ice is left skewed akin to cement-based materials. Hence, it can be inferred that the tangential/rolling bond strength envelope of MH resembles that of cement-based materials. Accordingly, the fitting parameters determined for the shape of the strength envelope in case of cement bonds [44] can be used for MH bonds. Thus, Equations (4) for MH bonds are here re-written as:

$$R_{sb0} = 1.38 \cdot 0.38 \cdot (F_n + R_{tb}) \cdot \left(\ln \frac{R_{cb} + R_{tb}}{F_n + R_{tb}} \right)^{0.59} , \qquad (19a)$$

$$R_{rb0} = 1.366 \cdot 0.741 \cdot (F_n + R_{tb}) \cdot \left(\ln \frac{R_{cb} + R_{tb}}{F_n + R_{tb}} \right)^{0.59} , \qquad (19b)$$

where R_{tb} and R_{cb} can be computed from a combination of Equations (12), (16) and (18) for a set of given parameters: T, σ_w, and ρ. According to Equations (12), R_{tb} and R_{cb} also depend on the level of MH saturation, S_{MH}, which rules the average width of bonds, B, hence the bond strength. The correlation between the S_{MH} and B will be further explained in Section 3.3. Figure 10 illustrates how the strength envelopes obtained for R_{sb0} and R_{rb0} vary with the level of MH saturation. The shape of the envelopes which is controlled by the fitting parameters remains unchanged with the variation of hydrate saturation, but the envelope size increases with the level of hydrate saturation. The relationship between hydrate saturation and the average width of bonds is further discussed in the next section.

3.3 Geometric features of inter-particle MH

From the experiments conducted by Hyodo et al. [32], it can be inferred that the inter-particle MH bond dimension varies with the level of MH saturation. As shown in Figure 2, a bond between cylindrical particles (or spherical ones in 3D) is geometrically described by its width, B, and its thickness at the center, t_0. It is convenient to define a dimensionless parameter β representing the ratio between bond and particle sizes:

$$\beta = B/\overline{R} \tag{20}$$

In the literature, the hydrate saturation degree, S_{MH}, is defined as the ratio of the methane hydrate volume to the total volume of the void. In the context of two-dimensional problems such as in this study, S_{MH} reduces to the ratio of the methane hydrate area A_{MH} (i.e., the area of void filled with bonds) to the total void area A_V, i.e., $S_{MH} = A_{MH}/A_V$. The area of void occupied by the i-th bond (the blue area as shown in Figure 2), A_{bi}, can be represented as a function of β from elementary trigonometric considerations assuming that the radii of the two bonded particles are equal to the equivalent radius, $\bar{R}$ (i.e. neglecting the different curvatures of the particles in the calculations):

$$A_{bi} = \bar{R}_i^2 \left[2\beta - \beta\sqrt{1-\frac{\beta^2}{4}} - 2\arcsin(\frac{\beta}{2}) + \frac{t_{0i}\beta}{\bar{R}_i} \right]. \tag{21}$$

The total area occupied by the bonds in a sample can be found by summation over all the bonds: $A_b = \sum_{i=1}^{m} A_{bi}$ with *m* being the total number of bonds, which depends on the initial configuration of the packing of the sample (i.e., whether loose or dense). Moreover, $A_{MH} = A_b + A_{MH_0}$ with A_{MH_0} being the area occupied by MH not binding any grains (i.e., floating around). Therefore, the level of hydrate saturation can be related to the area occupied by bonds as follows:

$$\begin{aligned} S_{MH} &= \frac{A_b + A_{MH0}}{A_V} = \frac{A_b}{A_V} + S_{MH_0} = \frac{(1+e_p)}{e_p A} A_b + S_{MH_0} \\ &= \frac{(1+e_p)}{e_p A} \sum_{i=1}^{m} \bar{R}_i^2 \left[2\beta - \beta\sqrt{1-\frac{\beta^2}{4}} - 2\arcsin(\frac{\beta}{2}) + \frac{t_{0i}\beta}{\bar{R}_i} \right] + S_{MH_0} \end{aligned} \tag{22}$$

where *A* is the total area of the sample; e_p is the planar void ratio; and S_{MH0} is the threshold value of hydrate saturation at which methane hydrates start to bond sand grains, which is around 20% - 30% [18, 19, 52, 53]. Note that the value of S_{MH0} depends on the hydrate growth history.

Equation (22) identifies a non-linear relationship that depends on the state of compaction of the sample (e.g., relative density), which rules the total coordination number of the sample and therefore the initial number of bonds, *m*. Figure 11 presents the relationship between $S_{MH} - S_{MH_0}$ and β achieved for the case of a dense sample (i.e., e_p= 0.21) at different critical values of bond thickness, t_0^{cr}, below which MH bonds form (i.e. bonds do not form for $t_0 > t_0^{cr}$). For an arbitrary value of t_0^{cr}, such relationship can be used to determine the value of β to be employed in the simulations for various levels of hydrate saturation S_{MH}.

Here, we want to replicate the natural process of formation of MH bonds as closely as possible. The natural process of reference is given by MH bonds formed by methane gas percolating through a deposit of sand particles. Hence, in our simulations, once the DEM sample has been isotropically compressed to the confining pressure of reference, bonds of various thickness were activated between particles with the minimum bond thickness t_0 (see Figure 2) coinciding with the gap (the minimum distance) between two adjacent particles.

It is important to note that MH bonds do not naturally exist in all the gaps of the sample. In fact if the distance between two adjacent particles is larger than the threshold value t_0^{cr}, there is little possibility for the formation of MH bonds. Figure 12(a) and (b) show typical SEM images of an artificial MHBS specimen with a MH saturation of 50% and its host sand, respectively. We made an estimate of the threshold distance t_0^{cr} based on several SEM images (as in Figure 12). After identifying the outlines of the MHs and the particles of the host sand, we measured the MH bond thickness t_0 at each particle contact. The measured value of t_0 was always lower than 5% of d_{50}, with d_{50} being the median particle diameter. Hence, the gap threshold for MH bond formation between two particles was taken as 5% of d_{50}. It is worthy to mention that the value of t_0^{cr} may depend on factors such as MH saturation, void ratio, type of host sand, which certainly need further investigation. Here, for the sake of simplicity, we chose the same value of t_0^{cr} (i.e., 5% of d_{50}) in all the simulations carried out.

Note that in the presented simulations once the bonds are broken they do not reform. This implies that the time scale of the loading is much higher than the time scale for bond formation so that MH induced bond formation during the occurrence of the tests is negligible. For the investigation of phenomena such as MH extraction induced submarine landslides and/or well instabilities, the assumption should be on the conservative side since the reformation of MH bonds is expected to have a stabilising effect opposing the development of failure mechanisms. However, in principle it is possible to consider bond reformation in the analyses provided that an evolution law for the bond strength over time, encapsulating the timescale of bond reformation, is assigned. Equally, any other time dependent phenomena occurring at the bond level could be considered by assigning a suitable time dependent bond law (Nicot, 2004a, b).

4 DEM biaxial tests on MHBS

In this section, the 2D - DEM biaxial tests carried out to investigate the macro-mechanical behaviors of MHBS are illustrated. Since in section 5 the DEM results have been compared with the experimental results conducted by Masui et al. [18], also a brief description of their test program is provided.

4.1 DEM simulation procedure

The proposed bond contact model for MHBS was compiled by C++ code and implemented into the commercial DEM software PFC2D [54] for the simulation of biaxial compression tests. The simulations were carried out in three steps: (1) generation of an un-bonded sample without cementation of MHs; (2) activation of MH bonds in the sample; and (3) shearing of the sample under a pre-selected confining pressure.

The ‘multi-layer with under compaction method’ proposed by Jiang et al. [55] was used to generate a homogenous un-bonded sample consisting of 6000 particles at a target void ratio of 0.21 which implies that the sample generated is dense. This sample was then isotropically subjected to a confining pressure of 1 MPa, the exact value applied in the tests conducted by Masui et al. [18]. Next, MH bonds were activated in the sample at the confining pressure of 1 MPa at a given environmental setting, i.e., surrounding temperature and backpressure. During the process of sample generation, the wall-particle friction was set to zero to eliminate any boundary effects. Figure 13 illustrates the DEM sample with a width of 400 mm and a height of 800 mm, in which the existence of the MH bonds is highlighted by black solid lines linking adjacent particles. The amplified part of the sample in Figure 13 shows that MH bonds have been successfully formed at all target contacts (i.e., whether virtual or real contacts).

After sample preparation, the sample was sheared under a constant strain rate of 5% per minute by moving the top and bottom walls. The confining pressure was kept constant at a pre-selected confining pressure during the shear phase by adjusting a numerical servo-mechanism to the side walls.

4.2 Sample parameters

Figure 14 provides the grain size distribution of the DEM sample in comparison to Toyoura sand adopted by Masui et al. [18]. The simulation used a more uniform sample with particle diameter ranging from 6.0 mm to 9.0 mm in order to achieve computational efficiency. The median diameter, d_{50}, of the DEM sample is 7.6 mm, and the uniformity coefficient is 1.3 (i.e., the ratio of d_{60}/d_{10}, with d_{60} or d_{10} being the particle diameter at which 60% or 10% of the mass of a soil sample is finer, respectively).

Table 4 lists the parameters employed in the simulation. In case of un-bonded particles, a trial-and-error procedure was used to determine the micro mechanical parameters. Parameter were chosen in order to match as closely as possible the material strength exhibited in triaxial tests on the host sand without any MHs.

In case of bonded particles, the parameters of the bond contact model (see Table 1) were determined via the relationships outlined in section 3 using as input the experimental conditions (temperature, pressure, MH density. Etc.) of the experiments run by Masui et al. [18]. The MH saturation degree used in the simulation was set to be 0%, 25%, 40% and 55%. The pore-filling part of MH saturation (i.e., S_{MH0} in Equation 22) was chosen as 20%. Accordingly, values of the bond area parameter β at different MH saturations were obtained by Equation (22) (see Table 4). In Figure 11 the relationship between parameter β and MH saturation is provided. Note that the relationship is specific to the PSD considered: if a different PSD is considered a different relationship has to be expected. The surrounding temperature and water pressure (i.e. backpressure applied to the sample) were +5^{0}C and 8.0 MPa, respectively. Assuming a typical value of MH density, 0.9 g/cm^3, the Young's modulus, the tension and compression strengths of MH were obtained by Equations (9), (16) and (18), and listed in Table 4. Based on the parameters listed in Table 4, the model parameters at each bond were computed according to Equations (6) and (12) by a C++ subroutine. In Figure 10a and b the envelopes representing the strength of the bond between two particles with average radius $\bar{R} = d_{50}/2$ subject to a tangential force and a rolling moment respectively for MH saturations of 25%, 40% and 55% are presented as an example. As shown in Figure 10, the value of all bond strengths (i.e. R_{tb}, R_{cb}, R_{sb0}, R_{rb0}) increases as the MH saturation increases, showing an enhancement of microscopic strength with MH saturation.

5. DEM Simulation results

5.1 Validation of the bond contact model

To check the validity of the bond contact model, numerically obtained macro-mechanical behaviors (e.g., the stress-strain behavior and the volumetric response) are presented here with respect to the MH saturation and effective confining pressure in comparison to the experimental results obtained by Masui et al. [18].

Figure 15 presents the simulated stress-strain response of MHBS at four different MH saturation degrees in comparison to the corresponding test results under the same conditions. The comparison shows that the numerical simulations can effectively replicate the influence of the MH saturation on stress-strain behaviors of MHBS for the following aspects: (1) strain softening appears and becomes more and more evident with the increasing of S_{MH}; (2) both the elastic modulus and maximum deviator stress increase gradually with the increasing of S_{MH} and the axial strain at the maximum deviator stress is around 2% ~ 3%, in good agreement with the experimental results; (3) the variation of deviator stress with S_{MH} decays with the axial stain after the deviator stress has reached the maximum value.

Based on the data in Figure 15, variations of the peak shear strength and the elastic modulus of MHBS with respect to the MH saturation degree, S_{MH}, are provided in Figure 16. Here, the peak shear strength is the deviator stress at its maximum value (i.e.,$\sigma_{peak} = (\sigma_1 - \sigma_3)_{\max}$), and elastic modulus, E_{50}, is the tangential modulus at the point where the deviator stress is one half of the peak value. As shown in Figure 16, the numerical results of the peak shear strength and elastic modulus increases gradually with S_{MH}, which qualitatively agrees well with the experimental data. Further, the variation of secant Young's modulus, E_{sec}, at various axial strains (e.g., 0.5%, 1% and 1.5%) with MH saturation obtained from the numerical results is presented in Figure 17. The secant modulus increases approximately linearly with S_{MH} at all axial strains. Intercept of the lines increases with the axial strain while no significant change is found in the slope. There is a large increase in E_{sec} at an axial strain of 0.5%, when S_{MH} changes from 0% to 25%, indicating that the presence of MH bonds greatly contributes to elastic modulus of the host sample, especially at small axial strains. This is also denoted in Figure 15(b) that the initial slope of the stress-strain relationship changes greatly when S_{MH} increases from 0% to 25%.

Figures 18(a) and (b) present the relationships between the volumetric strain and the axial strain obtained from the experiments and numerical simulations, respectively. It is clear that the volumetric strain in both numerical and experimental results exhibit initial contractive behavior followed by shear dilation. Moreover the shear dilation becomes evident with the increase of S_{MH} in both cases, showing a significant effect of MH bonds on the volumetric dilative response. The variation of dilation angle with S_{MH} is provided in Figure 19. The dilation angle in both experimental and numerical cases gradually increases with S_{MH}. In addition, Figure 19(a) provides the dilation angle obtained from numerical results at effective confining pressures of 2 MPa and 3 MPa in addition to 1 MPa. As shown in this figure, the dilation angle at the effective confining pressure of 2 MPa and 3 MPa are also enhanced by S_{MH}, while this angle decreases as the effective confining pressure increases.

Figure 20 provides an example of stress-strain behavior under different effective confining pressures obtained by experiments and simulations at similar S_{MH}. The stress-strain responses under these effective confining pressures exhibit strain-softening with the maximum deviator stress being greatly enhanced by the effective confining pressure. The variation of peak shear strength σ_{peak} with respect to the effective confining pressure is provided in Figure 21, in which the relationships with different S_{MH} are included. The peak shear strength increases dramatically as the effective confining pressure increases at any S_{MH}. The data in Figure 21 lead to a relationship between the peak strength parameters and S_{MH} as depicted in Figure 22.

In both numerical and experimental cases, the presence of MH causes considerable increase in cohesion, while no significant change is noted in the internal friction angle associated with the increasing of S_{MH}.

As discussed above, although the simulation cannot quantitatively reproduce the experimental tests, DEM implemented with the bond contact model can effectively capture the essential features relevant to the influence of the MH saturation degree and the effective confining pressure on the macro-mechanical behaviors of MHBS.

Constant volume (isochoric) tests were run for various values of the confining pressure and MH concentration to investigate the undrained behavior of the MHBS. As it is well known during the execution of an undrained test, excess pore pressures are generated so that a fully coupled liquid – solid numerical analysis would be necessary. However, (Shafipour and Soroush 2008) compared 2D fully coupled CFD – DEM analyses with 2D isochoric DEM analyses showing that constant volume DEM simulations are in good agreement so that it can be concluded that 2D isochoric DEM analyses may be used to investigate the material behavior in undrained conditions.

In figure xxx the results of undrained analyses run for various values of the confining pressure and MH concentration are shown. In all cases non liquefaction was exhibited. This is due to the fact that the samples considered were generated in loose conditions.

5.2 Microscopic information on bonds

Figure 23 provides the relationship between percentage of intact MH bonds and the axial strain (0~30%) obtained from the DEM tests on the MHBS with different MH saturation. Figure 23 shows that the percentage of intact bonds decreases with the axial strain at a decreasing rate. The intact bonds diminish remarkably in particular when the axial strain is lower 5%. This is the stage where the deviator stress drops from the peak to a relatively low value as shown in Figure 15(b). Figure 24 also presents the percentage of the intact bond area (i.e., A_{intact}/A_{total} , with A_{intact} being the area of the intact bonds and A_{total} being the total area of the MH bonds). As shown in Figure 23, the percentage of the intact bond area is a little higher than the percentage of the intact bond number. This indicates that the MH bonds with relatively lower area (i.e. the bonds with relatively lower width or between small particles) break more likely than others during the shearing process. This is reasonable since bonds with a lower width generally carry smaller bond strength.

Let us consider the sample with S_{MH} = 40% as an example to further clarify microscopic information on bonds. Figure 24 presents the stress-strain and volumetric strain response up to an axial strain of 30%, and the distribution of MH bonds at an axial strain of 2%, 5%, 15%. In Figure 24(b), only intact MH bonds are marked as black solid lines at particle contacts. Figure 24 shows that shear bands develop in the sample and most of the bonds break in the bands. The percentage of broken bonds increases with the expansion of shear bands when the axial strain grows. In addition, when the axial strain reaches 15%, there are still some intact bonds left in the sample (around 15% at S_{MH} = 40% as shown in Figure 23), and the intact bonded particles outside the bands act as clumps. As a result, these intact bonds are expected to break only if very large deformation of the sample occurs (e.g., 7% for S_{MH}=40% at axial strain of 30% as shown in Figure 23). As indicated by Figure 24(a), the breakage of remaining bonds contributes little to the deviator stress and volumetric strain of MHBS when the axial strain reaches 15%. Hence, it can be concluded that the critical state in terms of conventional definition can be reached while the bonds are not fully broken. This may explain that the strength of the host sand coincides nearly with the strength of the MH bearing sand in real experiments at an axial strain of around 15% as shown in Figure 15.

Figure 25 presents the contact force chains for samples with MHs (S_{MH} = 40%) and without MHs at the axial strain of 0% (onset of the shear phase), 2% (the peak deviator stress) and 15% under the effective confining pressure of 1 MPa. The thickness of the force chains is proportional to the value of contact forces, while the direction represents the direction of the contact force. Figure 25 shows that the contact force chains of the sample with MHs resemble to those without MHs at the onset of the shear phase. This is because that the sample without activating the MH bonds had reached the equilibrium under the confining pressure of 1 MPa. During the process of bond activation, particles remained steady under such stress state and no significant extra forces were resulted due to bond activation. However, the stress state of the samples changes gradually due to the vertical loading during the shear process. The contact force chains exhibit differently in two cases, because MH bonds can bear additional inter-particle forces caused by loading than contacts without bonds. As shown in Figure 25 (a) and (b), tension forces marked as the red lines appear in the samples with MHs, while only compression force are found at the particle contacts in the samples without MHs. Besides, the compression force of the samples with MHs is larger than that without MHs due to the contribution of MH bonds. When the axial strain increases to 15%, the contact force chains of sample with MH bonds resemble to those without MH bonds. This further indicates that the strength of the MHBS tends to be close to the strength of the host sand associated with the increasing of axial strain due to the large number of bond breakage (see Figure 24).

5.3 Discussion

The DEM simulation incorporating the bond contact model of MHBS can capture several key features of MHBS, particularly the bonding effects of MHs, providing an effective tool to understand the micro-mechanism of the mechanical behaviors of MHBS. However, some differences were identified between the numerical and the experimental data [18].

A 2D model was used in the present study instead of 3D models due to several reasons. First of all there is no available experimentally-validated contact law for three-dimensional cemented granules. Many efforts were made in the past several years to develop reliable equipment and repeatable procedure for conducting a micro mechanical experiment to validate the bond contact law [43, 44, 56]. However this work was only restrained to 2D context using rods, because the test on spheres in three dimensions will become extremely difficult in particular when a complex of loads is of desire. Second, DEM simulations of engineering problems of interest, e.g., landslides along the seabed or well instabilities induced by MH extraction, are computationally very expensive and hence are only possible in 2D for the foreseeable future. Third, though 3D modeling can provide more realistic results, the 2D model can be still used as a useful research tool due to simplicity in implementation and easy interpretation of results. Moreover, 2D simulation can still shed light on the importance of MH bonds in MHBS and the growing pattern of shear bands in MHBS, which will not significantly differ from that in 3D modeling. Although 2D modeling has limit to quantitatively replicate volumetric responses obtained from the 3D triaxial tests, it can still produce realistic strength parameters, particularly the friction angle, which has been demonstrated in Figure 22.

Granules of the host sand were idealized as disc-shaped particles, which differ from the realistic soil gains with irregular shapes. Excessive rotation will be resulted from circular particles if no particular action is taken to improve the contact model. In order to imitate the realistic shapes of soil grains and, more importantly, produce realistic bulk strength of soils, a few techniques are now available, such as clustered discs [57], polygon [58], polyarc elements [59] among others (e.g., [60]), all of which can reduce excess particle rotation. However, using particles with irregular shapes will give rise to some practical issues in addition to the loss in computational efficiency due to demand for a rigorous contact detection algorithm and others. For instance, extreme effort will be certainly needed to accurately generate even a small sample with exact microstructure (e.g., shape and fabric) as real soils. Moreover, particle angularity will evolve during loading process, and such morphological change becomes almost untraceable in a practical simulation using particles of sufficient number. Alternatively, realistic particle rotation can still be achieved from disc-shaped particles by directly incorporating the inter-particle rolling resistance into the contact law in the manner that this present study adopted (e.g., [45, 61, 62]). This approach sacrifices details at the particle scale such as the particle shape, but is particularly suitable where accurate bulk behaviors of the medium are prior to fidelity to the actual particle shapes.

The grain size distribution of the DEM sample differs from that of Toyoura sand used in the experiment of Masui et al. [18]. The average particle diameter of the DEM sample (i.e., 7.6 mm) is larger than that of Toyoura sand (i.e., below 0.2 mm). Larger particles were used in this study for computational efficiency. As long as the model parameters are well calibrated following an appropriate procedure, an assemblage of large particles can still replicate realistic mechanical properties of soils, which is of the most value among other particle characteristics for the goal of this paper. This has been demonstrated in Figures 15 to 22. Note that this bond contact model and the procedure for determining the model parameters presented here are also applicable to MHBS samples with finer particles except that the best-fit parameter set may be different from the one presented herein due to the differences in the particle size.

This work has a few limitations. The focus of this study was only on the MHs acting as the cementation agent at inter-particle contacts in MHBS. The contribution from other types of MHs, such as pore-filling and coating, may be under- or over-estimated. An arbitrary value was assigned to the threshold value of the hydrate saturation degree, below which only pore-filling MHs will form and this type of MHs is assumed to carry negligible effects on the bulk mechanical properties of MHBS. However, pore-filling MHs may have some effect on the strength of MHBS since they can bear and transmit loads as the soil skeleton deforms. This kind of effect was revealed by Brugada et al.[8]. It is noted that simultaneous consideration of different types of MHs will become meaningful in DEM analyses only if identification and quantification of different types of MHs become possible in experiment with the advancement of microstructure detection techniques.

6 Conclusions

This study proposed a two-dimensional bond contact model for the DEM simulation of soils with methane hydrates (MHs) cementing at inter-particle contacts of grains. A conceptual bond contact model [36], which has been validated through a specifically-designed test [43, 44], was first introduced with an emphasis on the generic contact law, which was later adapted in MHBS. Due to difficulty in conducting the micro-mechanical tests on bonded granules using real MHs, the model parameters in relation to the bond strength and particle stiffness were suggested to be formulated from the correlation to the surrounding temperature, water pressure and MH density. The bond width and thickness varying at each bond were related to a pre-selected value of MH saturation degree. With the aid of a customized C++ subroutine, the proposed model was successfully implemented into a DEM software, PFC2D, and then testified through a series of biaxial compression tests on MHBS at various levels of MH saturation and different surrounding environmental conditions (i.e., different temperatures and pressures).

Compared with the experimental data obtained by Masui et al. [18], the DEM simulation can effectively capture the major mechanical behaviors of MHBS, e.g., the phenomenon of strain softening and shear dialation which become evident with the increasing of the hydrate saturation degree. In particular, the model is able to reproduce realistic strength properties of MHBS samples at a wide range of the hydrate saturation degree, which is in good agreement with the experimental results. Although some quantitative differences with the experimental triaxial tests were observed, the proposed bond model and its parameter calibration proved to be realistic so that the proposed bond model can be employed for DEM simulations of problems of engineering interests.

The proposed model can also serve as a useful tool to better understand the connection between the microscopic behaviors and the macroscopic properties of MHBS samples. The presented set of simulations shed light on the mechanical behavior of the MH bearing sands at the microscopic level, e.g., spatial distribution of bond breakage, type of bond failure (either shear or tensile), redistribution of forces within contact chains so that our understanding is clearer on the relationship between cementation and de-bonding mechanisms occurring at the micro level and macro-mechanical behavior of MHBS.

Acknowledgments

This research was financially supported by China National Funds for Distinguished Young Scientists (Grant No. 51025932), National Natural Science Foundation of China (Grant No. 51239010), and Ph.D Programs Foundation of Ministry of Education of China with (Grant No. 20100072110048). These joint supports are sincerely appreciated.

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