DEM simulations of methane hydrate exploitation by thermal recovery and depressurization methods

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Abstract. Methane hydrate (MH, also called fiery ice) exists in forms of pore filling, cementing and load-bearing skeleton in the methane hydrate bearing sediment (MHBS) and affects its mechanical behavior greatly. To study the changes of macro-scale and micro-scale mechanical behaviors of MHBS during exploitation by thermal recovery and depressurization methods, a novel 2D thermo-hydro-mechanical bonded contact model was proposed and implemented into a platform of distinct element method (DEM), PFC2D. MHBS samples were first biaxially compressed to different deviator stress levels to model different in-situ stress conditions. With the deviator stress maintained at constant, the temperature was then raised to simulate the thermal recovery process or the pore water pressure (i.e. confining pressure for MH bond) was decreased to simulate the depressurization process. DEM simulation results showed that: during exploitation, the axial strain increased with the increase of temperature (in the thermal recovery method) or decrease of pore water pressure (in the depressurization method); sample collapsed during MH dissociation if the deviator stress applied was larger than the compression strength of a pure host sand sample; sample experienced volume contraction but its void ratio was slightly larger than the pure host sand sample at the same axial strain throughout the test. By comparison with the laboratory test results, the new model was validated to be capable of reproducing the exploitation process by thermal recovery and depressurization methods. In addition, some micro-scale parameters, such as contact distribution, bond distribution, and averaged pore rotation rate, were also analyzed to investigate their relationships with the macroscopic responses.

Keywords: methane hydrate; distinct element method; thermal recovery; depressurization; macro- and micro mechanical responses

1. Introduction

A methane gas molecule (CH_4) can be caged by water molecules under high pressure and low temperature conditions [1-4]. The aggregation of such cages is an ice-like crystal called methane hydrate (MH) with properties similar to ice [5]. As found in previous investigations [6], a great amount of MH exists stably undersea. The mixture of MH and seabed sediment is called methane hydrate bearing sediments (MHBS).

MH exploitation has attracted great attentions since the total carbon content in MH is twice as large as that in petroleum, coal and methane gas available now [7]. Moreover, it is considered as a type of promising and hugely reserved clean energy which can alleviate the energy crisis to certain extent [8-11]. Therefore, how to exploit MH safely and efficiently has become a worldwide focus [12].

The exploitation methods mainly include the chemical and physical methods. For chemical methods such as chemical injection method, the diffusion of chemicals is inhibited due to the low permeability of MHBS [13], leading to low gas production rate. Moreover, the costs of chemical method are high and not suitable for large scale exploitation [14, 15]. These two defects make researchers seek other more efficient methods. Physical exploitation methods, mainly including the thermal recovery and depressurization methods, have the advantages including low costs, high gas productivity, etc. [16, 17]. The principle of these two methods is to change the occurrence environment of MH by either increasing the temperature or reducing the confining pressure until the temperature-confining pressure condition is brought out of the stable zone and MH dissociates into methane gas and water [18, 19]. Despite the obvious advantages of physical methods, some accidents happened in these processes such as submarine landslides, platform foundation settlement and failures of lifeline engineering projects [20-22].

To study the dissociation process, previous in-situ tests [23, 24] have only investigated the change of modulus by measuring the shear wave propagation velocities through the MHBS, while other mechanical properties, such as stress-strain relationships, could not be studied during the in-situ MH dissociation. It is not practical to carry out laboratory tests either, because the occurrence conditions of MH are too difficult to maintain and samples are easily disturbed during transportation. Therefore undisturbed samples are difficult to attain for laboratory tests [25, 26] and many previous laboratory tests were performed with artificial samples. For example, Lee et al.

[27] studied the effect of MH saturation on volumetric changes during MH dissociation. The results have shown that there is a volumetric contraction regardless the initial MH saturation and the contraction ratio increases with the increase of saturation. Hyodo et al. [28] investigated the changes of MHBS mechanical properties during MH dissociation using an improved triaxial apparatus. The results indicate that dissociation can lead to volumetric contraction, and the stress-strain relationship depends on the deviator stress applied. Laboratory tests can provide useful information for understanding the mechanical process of dissociation and improving the MH exploitation method. However, many challenges were encountered in experiments, such as difficulties in replicating samples with the same physical properties or visualizing the changes of microscopic structure, which is believed to underlie the macroscopic behaviors.

The distinct element method (DEM) [29], which was developed on the basis of simple inter-particle contact laws, has advantages of visualizing the evolution of microscopic particle interactions with good sample repeatability and low costs. It has attracted many interests in the geotechnical community [30-32]. Holtzman et al. [33] simulated the dissociation of MHBS by deleting small particles, and simulated the change of pore water pressure by changing the effective confining pressure. Test results have shown that the elastic modulus decreases with increasing MH saturation or decreasing excess pore water pressure. This simulation demonstrated that DEM could be used to simulate the dissociation process of MH in MHBS.

Previous studies have shown that the MH bonded with soil particles has larger effects on mechanical properties of MHBS than other types of MH [34-36], therefore the aim of this paper is to study the effects of bonded MH. For this purpose, a bonded contact model is required. Jiang et al. [37, 38] has proposed a bonded contact model calibrated from a large amount of experimental results of two aluminum rods bonded by cement or epoxy resin. However, this model did not consider the bond thickness, which was affected by hydrate saturation and could affect the bond strength. The coupled effect of temperature and confining pressure on the bond strength was not considered either. This paper first proposed a new model with consideration of the effect of bond thickness, temperature and confining pressure. This model was then implemented into a DEM platform, PFC2D, to simulate the MHBS exploitation by thermal recovery and depressurization. DEM simulation test results were then compared with laboratory tests conducted by Hyodo et al. [28] to validate the new model.

2. A New bonded contact model for MHBS

To capture the mechanical behaviour of MH bond, the existing contact- and parallel-bond model in PFC2D was developed further by implementing the previous experimental findings of pure methane hydrate. The contact behavior between two particles and the bond before bond breakage is firstly introduced in Section 2.1. In this section, what the authors defined directly is the strength and stiffness of pure methane hydrate bond neighboring two particles, rather than the strength and stiffness of MHBS. The MH bond breakage criterion, as described in Section 2.2, is dependent on the MH bond stiffness and MH bond strength. Sections 2.3 and 2.4 detail the developments of empirical models for MH bond stiffness and MH bond strength, respectively, based on collections of previous experimental data, including the key data from the triaxial tests on pure MH samples performed by Hyodo et al. [19]. In MHBS, the pores are not only filled by MH bond but also by pore water, and solid MH bond is surrounded by water. To simulate the MHBS exploitation, each MH bond at contact is idealized as a pure MH sample subjected to a triaxial test, where the pore pressure (back pressure) in the MHBS sample is idealized as the “confining pressure” on the pure MH sample equivalent to an experimental test performed by Hyodo et al. [19].

Fig. 1. Schematic diagram of connection between lab test of methane hydrate and DEM bond model for simulations of MHBS

2.1 Contact behavior before bond breakage

There are two types of contacts as shown in Fig. 2. In Type 1, two soil particles with radii of R_1 and R_2 are in separation when the bond forms. The MH bond has a width of B , a maximum thickness $h_{\max}^{cr}$ and a minimum thickness l ($l > 0$), as shown in Fig. 2(a), i.e. $u_n = R_1 + R_2 - d = -l < 0$,

where d is the distance between the centers of two bonded particles. In Type 2, two particles are in contact when the MH bond forms, i.e. $u_n \geq 0$, as shown in Fig. 2(b). **Note that although not illustrated in Fig. 2 for readability, MH bond in these two types of contact are exposed to water pressure directly in MHBS as shown in Fig.1 since MH bond is surrounded by pore water.**

The formation of MH bond is determined by two factors. One is the maximum separation between two particles, because MH bonds can only form with the thickness not larger than a critical distance $h_{\max}^{cr}$. The other factor is MH saturation, which is defined as the ratio of MH area over the total void area in the two-dimensional case. It determines the critical distance $h_{\max}^{cr}$. As shown in previous studies [39, 40], MH exists not only in the form of bond, but also in other forms such as pore filling or particle coating. When the saturation is less than 20%, MH exists in the forms of pore filling or particle coating; when the saturation is larger than 20%, MH bond starts to form. Thus, MH saturation S_{MH} can be expressed as

$$S_{MH} = \frac{A_b}{A_v} + S_{MH0} \quad (1)$$

where A_b is the total area of MH bond, and A_v is the total void area. S_{MH0} is the critical saturation that MH bond starts to form. S_{MH0} was taken as 20% in these study [39, 40]. A_b can be expressed as

$$A_b = \sum_{i=1}^m A_{bi} \quad (2)$$

where m is the total number of MH bonded contact and A_{bi} is the MH bond area at contact i , which can be expressed as :

$$A_{bi} = h_{\max}^{cr} B_i - 2\bar{R}_i^2 \arctan \frac{B_i}{\sqrt{4\bar{R}_i^2 - B_i^2}} + B_i \sqrt{\bar{R}_i^2 - \left(\frac{B_i}{2}\right)^2} \quad (3)$$

where $\bar{R}_i = 2R_{1,i}R_{2,i} / (R_{1,i} + R_{2,i})$ with $R_{1,i}$ and $R_{2,i}$ being the radii of two particles bonded by MH bond at contact i ; $B_i = \sqrt{4\bar{R}_i^2 - (2\bar{R}_i + l_i - h_{\max}^{cr})^2}$ is the bond width. It can be determined if $h_{\max}^{cr}$ is given. The relationship between S_{MH} and $h_{\max}^{cr}$ is shown in Fig. 3.

Fig. 2. Cemented hydrates at inter-grain contacts

Fig. 3. Relationship between S_{MH} and h_{max}^{cr}

Fig. 4 illustrates the contact force transmission mechanisms for the two different types of bonded contacts. Forces and moments in type 1 contact are all transmitted through MH bond as shown in Fig. 4(a), which can be expressed as

$$F_n = F_n^b, F_s = F_s^b, M = M^b \quad (4)$$

where F_n , F_s and M are the total normal force, shear force and moment transmitted through the contact, respectively; F_n^b , F_s^b and M^b are normal force, shear force and moment transmitted through MH bond, respectively.

The force-displacement relationship of MH bond can be expressed as

$$F_n^b = k_n^b \cdot u_n^b, \quad -R_{tb} < F_n^b < R_{cb} \quad (5)$$

$$F_s^b \leftarrow k_s^b \cdot \Delta u_s^b + F_s^b, \quad F_s^b < R_{sb} \quad (6)$$

$$M^b \leftarrow k_m^b \cdot \Delta \theta^b + M^b, \quad M^b < R_{rb} \quad (7)$$

where R_{tb} , R_{cb} , R_{sb} and R_{rb} are tension, compression, shear and rolling strengths of MH bond, respectively; k_n^b , k_s^b and k_m^b are the normal, shear and rolling stiffnesses of MH bond,

respectively; u_n^b is the normal displacement of MH bond which can be expressed as $u_n^b = u_n - u_{n0}$, where u_{n0} is the initial overlap between particles when a MH bond forms; Δu_s^b and $\Delta \theta^b$ are the incremental relative shear displacement and rotational angle following MH bond formation, respectively.

Forces and moments in type 2 contact, as shown in Fig. 4(b), are transmitted through both MH bond and physical inter-particle contact, which can be expressed as

$$F_n = F_n^b + F_n^p, F_s = F_s^b + F_s^p, M = M^b + M^p \quad (8)$$

where F_n^p , F_s^p and M^p are normal force, shear force, and moment transmitted between particles, respectively. The force-displacement relationship of particles can be expressed as

$$F_n^p = \begin{cases} k_n^p \cdot u_n, & u_n \geq 0 \\ 0, & u_n < 0 \end{cases} \quad (9)$$

$$F_s^p \leftarrow \min[k_s^p \Delta u_s^p + F_s^p, \mu^p F_n^p] \quad (10)$$

$$M^p \leftarrow \min[k_m^p \Delta \theta^p + M^p, \frac{F_n^p \beta^p \bar{R}}{6}] \quad (11)$$

$$k_m^p = k_n^p (\beta^p \bar{R})^2 / 12 \quad (12)$$

where k_n^p , k_s^p and k_m^p are the normal, shear and rolling stiffnesses of particles, respectively; Δu_s^p and $\Delta \theta^p$ are the incremental relative shear displacement and rotational angle, respectively; μ^p is the inter-particle friction coefficient; β^p is the inter-particle rolling resistance coefficient [41].

Fig. 4. Interaction transmission in two type of contacts

2.2 MH bond breakage criterion

Laboratory tests have been conducted on pairs of rods bonded by cement or epoxy resin to study the bond failure criteria [37, 42-43]. A bond failure law has been proposed as

$$\left(\frac{F_s^b}{R_{sb}'}\right)^2 + \left(\frac{M^b}{R_{rb}'}\right)^2 \begin{cases} < 1 & \text{intact bond} \\ \geq 1 & \text{failure} \end{cases} \quad (13)$$

$$R_{sb}' = \mu^b \cdot R_{cb} \cdot \frac{F_n^b + R_{tb}}{R_{cb} + R_{tb}} \cdot [1 + g_s \cdot (\ln \frac{R_{cb} + R_{tb}}{F_n^b + R_{tb}})^{f_s}] \quad (14)$$

$$R_{rb}' = \frac{R_{cb} \beta^b \bar{R}}{6} \cdot \frac{F_n^b + R_{tb}}{R_{cb} + R_{tb}} \cdot [1 + g_r \cdot (\ln \frac{R_{cb} + R_{tb}}{F_n^b + R_{tb}})^{f_r}] \quad (15)$$

where R_{sb}' is the shear strength under tension-compression-shear condition; R_{rb}' is the rolling strength under tension-compression-torsion condition; μ^b is the friction coefficient of broken bond; β^b is the rolling resistance coefficient which can be referred to Jiang et al. [43]; f_s , g_s , f_r , g_r are fitting parameters for R_{sb}' and R_{rb}' , which are dependent on bond thickness and given by:

$$g_s = 2.876 - 1.623e^{-0.5((l-1.236)/0.506)^2} \quad (16)$$

$$f_s = 0.824 + 0.364e^{-0.5((l-1.069)/0.353)^2} \quad (17)$$

$$g_r = 3.068t^2 - 7.347l + 6.358 \quad (18)$$

$$f_r = 1/(2.719 - 3.207l + 1.442l^2) \quad (19)$$

The failure envelope can be visualized as shown in Fig. 5.

Fig. 5. Failure envelope of MH bond

Once the MH bond is damaged, the force-displacement law is replaced by a frictional rolling resistance law, which can be expressed as

$$F_n^b = \begin{cases} 0, & u_n^b < 0 \\ k_n^b \cdot u_n^b, & 0 \leq u_n^b < R_{cb}/k_n^b \\ \lambda R_{cb}, & u_n^b \geq R_{cb}/k_n^b \end{cases} \quad (20)$$

$$F_s^b = \min[k_s^b \cdot \Delta u_s^b + F_s^b, F_{s, resid}^b] \quad (21)$$

$$M^b = \min[k_m^b \cdot \Delta\theta^b + M^b, M_{resid}^b] \quad (22)$$

where λ is a parameter describing residual bond compression strength; $F_{s,resid}^b$ is the maximum shear strength of residual MH bond, which can be expressed as $F_{s,resid}^b = F_n^b \cdot \mu^b$; M_{resid}^b is the maximum residual rolling strength, which can be expressed as $M_{resid}^b = F_n^b \cdot B / 6$.

In summary, there are seven independent parameters (R_{tb} , R_{cb} , k_n^b , k_n^p , μ^b , μ^p , β^p) in the new bonded contact model, and three of them (R_{tb} , R_{cb} , k_n^b) are affected by temperature and confining pressure. Determinations of these three parameters are described in the following Sub-sections.

2.3 MH bond stiffness

Previous studies have showed that temperature, confining pressure and MH density affect the mechanical behavior of MH [19, 44-48]. In order to capture the coupled effects of temperature and confining pressure, a distance parameter L , which is defined as the shortest distance between the normalized test condition point and the normalized stability boundary line as shown in Fig. 6(b), was introduced. The normalized stability boundary line was calibrated from the stability boundary line proposed by Hyodo et al. [44]. It was probably first found in [44] that L is a suitable intermediate parameter to link methane hydrate strength and temperature/confining pressure in a simple linear way with satisfactory correlation. It is not necessary but indeed it is convenient to summarize the effect of two variables into one. This is due to the same mechanism in terms of methane hydrate crystal structure strengthening behind the effects of temperature and confining pressure. Any other parameter may be defined as well as long as it is easy to use and it has some physical meaning. If the test condition is in the upper left zone to the stability boundary line, MH is stable. With L increasing (temperature decreasing and confining pressure of MH increasing), MH strength and modulus increase as the cage type skeleton of methane hydrate becomes more stable; when the test condition is very close to the stability boundary line, i.e., L is near zero, the compressive strength of MH is almost equal to that of an ice as the cage type methane hydrate skeleton is unstable [45]. If the test condition is in the lower right zone to the stability boundary line, MH would dissociate.

Fig. 6 also presents various test conditions attained from previous experimental triaxial tests on

pure MH samples [19, 44-48]. It demonstrated that MH existed stably in the upper left zone to the stability boundary line.

The normalized temperature and confining pressure are defined as $T^*=T/T_r$ and $\sigma_w^*=\sigma_w/\sigma_{wT}$, where T_r and σ_{wT} are reference temperature and reference confining pressure. To determine T_r and σ_{wT} , 30 pairs of T_r and σ_{wT} were attempted, with T_r in the range of (235K, 285K) at an interval of 10K and σ_{wT} in the range of (1MPa, 13MPa) at an interval of 3MPa. All the previous stable test conditions as shown in Fig. 6(a) were normalized by each pair of T_r and σ_{wT} , and a distance parameter L for each test condition was then calculated. A linear regression model between the peak deviator stress $q_{\max,c}$ and the distance parameter L was established for each pair of T_r and σ_{wT} . The final T_r and σ_{wT} was selected as 235K and 1MP, as shown in Fig. 7, as their correlation coefficient R^2 of the linear regression was largest (0.92).

(a)

(b)

Fig. 6. Stability boundary line of MH under various test conditions (a) before normalization; (b) after normalization

Fig. 7. Relation between L and $q_{\max,c}$

To calculate the stiffness of MH bond (k_n^b), the MH bond was divided into two parts as shown in Fig. 8. Part I with normal contact stiffness k_{n1}^b is a rectangle with a width of B and a height of l (the minimum thickness of MH bond); part II with normal contact stiffness k_{n2}^b is similar to type 2 bonded contact with a minimum bond thickness of zero and a maximum thickness of $(h_{\max}^{cr} - l)$. The resultant normal stiffness of MH bond is a result of series connection of the two parts, and can be expressed as

$$k_n^b = \frac{k_{n1}^b \cdot k_{n2}^b}{k_{n1}^b + k_{n2}^b} \quad (23)$$

k_{n1}^b can be expressed as

$$k_{n1}^b = \frac{E \times l}{B} \quad (24)$$

where E is the elastic modulus of MH.

For part II, MH bond was divided into ten stripes with a width of $B/10$, and each stripe was approximated as a rectangle. The normal stiffness can thus be expressed as

$$k_{n2}^b / (E \times l) = \sum_{i=1}^{10} \frac{\Delta B^i}{(h^i - l)} \quad (25)$$

where h^i and ΔB^i are the height and width of stripe i , and $\Delta B^i = B/10$.

Fig. 8. Schematic diagram of MH bond for stiffness calculation

The shear stiffness of MH bond can be expressed as [41]

$$k_s^b = k_n^b / 1.5 \quad (26)$$

The rolling stiffness of MH bond can be expressed as [41]

$$k_m^b = \frac{k_n^b B^2}{12} \quad (27)$$

In summary, there is only one parameter E remained to be determined as the bond size can be determined as described in Section 2.1. E is related not only with temperature and back pressure, but also with MH density [19, 46]. The relationship between E and distance parameter L as well as MH density ρ^* ($\rho^* = \rho / \rho_{w,4^\circ\text{C}}$, ρ - density of MH; $\rho_{w,4^\circ\text{C}}$ - water density at a temperature of 4 °C) as established by Hyodo et al. [19] based on the experimental results of sample b with high MH content was adopted and implemented into the current bond model. The relationship can be illustrated in Fig. 9 and expressed as

$$E / p_a = L \cdot \beta_1 + \beta_2(\rho^*) \quad (28)$$

where β_1 is the slope and β_2 is the intercept, which is a function of ρ^* ; p_a is the standard atmosphere (1.01×10^5 Pa).

Fig. 10 shows the relationship between $\beta_2(\rho^*)$ and ρ^* [19]. Incorporating both, the elastic modulus E of MH can be expressed as

$$E / p_a = \begin{cases} 7840L + 8620\rho^* - 4890 & L > 0 \\ 0 & L \leq 0 \end{cases} \quad (29)$$

Fig. 9. Relation between E and the distance parameter L [19]

Fig. 10. Relationship between β_2 and ρ^* [19]

2.4 MH bond strength

MH bond compression strength R_{cb} and tension strength R_{tb} can be expressed as

$$R_{cb} = 1 \times B \times q_{\max,c} \quad (30)$$

$$R_{tb} = 1 \times B \times q_{\max,t} \quad (31)$$

where 1 means a unit length perpendicular to the deformation plane; $q_{\max,c}$ is the peak compression stress; $q_{\max,t}$ is the peak tension stress. They can be determined as follows.

Similar to the calculation of MH elastic modulus, MH strength can also be related to temperature, confining pressure and MH density. Fig. 11 presents the relationship between L and the peak deviator stress of sample b tested in [19]. By linear fitting of the experimental data, a relationship between the peak deviator stress of MH and L can be expressed as

$$q_{\max,c} / p_a = \alpha_1 L + \alpha_2(\rho^*) \quad (32)$$

where α_1 is the slope, and α_2 is the intercept which is a function of ρ^* .

Fig. 11. The relationship between L and the peak deviator stress of Sample b [19]

Fig. 12 shows the relationship between $q_{\max,c} / p_a$ and ρ^* . It shows that $q_{\max,c} / p_a$ increases linearly with MH density and confining pressure. These three lines under different confining pressures are nearly parallel. Thus, it is assumed that the slope is not affected by confining pressure. The relationship between $q_{\max,c} / p_a$ and ρ^* thus can be expressed as

$$q_{\max,c} / p_a = 186\rho^* + D(L) \quad (33)$$

where $D(L)$ is a function of L . Note that the difference between the solid line and the dash line in Fig. 12 is very small compared with the value of $q_{\max,c} / p_a$.

Fig. 12. Relationship between $q_{\max,c}/p_a$ and ρ^* [19]

MH dissociates if the test condition point is out of the stable zone, i.e., $L < 0$. Incorporating Eq. (32) and Eq. (33) we can get

$$q_{\max,c} / p_a = \begin{cases} 186\rho^* + 715L - 133 & L > 0 \\ 0 & L \leq 0 \end{cases} \quad (34)$$

Since there is no laboratory test determining the MH tension strength, in this paper, a hypothesis was proposed that if the compression strength is σ_w ($\sigma_w = \sigma_t + q_{\max,t}$) with the confining pressure of σ_t in a triaxial compression test, then the tension strength will be σ_t ($\sigma_t = \sigma_w - q_{\max,t}$) with the confining pressure of σ_w in a triaxial tension test. Note that the confining pressure for a MH bond is the pore water pressure in the MHBS sample as the MH bond is surrounded by pore water. Thus $q_{\max,t}/p_a$ can be expressed as

$$q_{\max,t} / p_a = \begin{cases} 186\rho^* + 715L(\sigma_t, T) - 133 & L > 0 \\ 0 & L \leq 0 \end{cases} \quad (35)$$

The validation of this model of MHBS without dissociation is in reference [49].

3 DEM simulations of thermal recovery and depressurization

3.1 Sample preparation

In order to validate the newly proposed MH bond model, thermal recovery and depressurization processes were simulated on MHBS samples under the same test conditions as those in [28]. In a MHBS sample, sand particles were modelled as 2D disks and MH bond were simulated virtually by specifying bonding parameters as described in Section 2. In all DEM simulations, no pore water was simulated. The biaxial compression test was performed by maintaining the effective confining pressure (σ'), i.e. the balance between the total confining pressure and the back pressure (pore water pressure, σ_w).

Using PFC2D, samples consisting of disks were prepared using the Multi-layer Under-compaction Method (UCM), which was first proposed in [50]. A granular sample with particle size distribution as shown in Fig. 13 was used herein. Particle diameter ranges from 6.0 mm to 9.0 mm with a mean diameter $d_{50}=7.6$ mm and a uniformity coefficient $C_u = 1.3$. The sample size is 800 mm $\times$ 400 mm with 6000 particles. Parameters adopted in the DEM simulations are shown in Table 1.

Using the UCM method, particles were compressed layer by layer to a target void ratio of $e_p=0.25$ first. Then an effective confining pressure $\sigma'=0.2$ MPa was applied isotropically until the

Fig. 13. Particle size distribution

Table 1. Particle parameters in DEM simulation

Particle	Sample size	800mm $\times$ 400mm
	Particle number	6000
	Particle density	2600kg/m ³
	Normal stiffness k_n^p	6.0×10^8 N/m
	Tangential stiffness k_s^p	4.0×10^8 N/m
	Friction coefficient μ^p	0.5
	Rolling coefficient β^p	0.5
Frictionless boundary	Friction coefficient μ	0.0
	Normal stiffness	6.0×10^8 N/m
Void ratio before consolidation		0.25

pure sand sample reaches equilibrium. MH bonds were then formed with a MH saturation of 50%

pure sand sample reaches equilibrium. MH bonds were then formed with a MH saturation of 50% ($h_{\max}^{cr}$ is found to be 1.2mm from Fig. 3). Parameters of MH bond (E , $q_{\max,c}$ and $q_{\max,t}$) were determined by Eqs (29), (34) and (35) given the initial temperature $T=278$ K and confining pressure for MH bond $\sigma_w=10$ MPa [28]. Thus R_{tb} , R_{cb} and k_n^b can be determined from Eqs (30), (31) and (23), respectively. The effective confining pressure σ' was then increased from 0.2MPa to 5MPa by moving boundaries to simulate the submarine condition as achieved in [28], i.e. the total confining pressure at 15MPa and the back pressure (i.e. pore water pressure and also confining pressure for MH bond, σ_w) at 10MPa. Hereafter the effective confining pressure σ' was maintained at 5MPa. Three deviator stresses of 0MPa, 3.8MPa and 6.8MPa were applied, equivalent to those in [28], corresponding to deviator stresses being zero, smaller than the strength of pure sand and larger than the strength of pure sand, respectively.

3.2 Test condition

In the laboratorial thermal recovery process [28], temperature was raised while the back pressure and deviator stress were maintained at the values described above. Note that confining pressure was not affected by methane gas generation as the exhaust condition of this sample was good. In the depressurization test [28], deviator stress and temperature were remained constant while the back pressure decreased from 10MPa to 3.5 MPa firstly to allow MH dissociate and then increased back to 10MPa to simulate the injection of pore water after dissociation. Changes of temperature and back pressure in the laboratory tests are shown in Fig. 14. These changes can be approximated by the mathematic expressions as:

$$T = \begin{cases} 10t + 278, & t \leq 1 \\ 5t + 283, & 1 < t \leq 2 \\ 293 & 2 < t \leq 4 \end{cases} \quad (36)$$

$$\sigma_w = \begin{cases} 10, & t = 0 \\ 3.5, & 0 < t \leq 5 \\ 6.5t - 29 & 5 < t \leq 6 \\ 10 & t > 6 \end{cases} \quad (37)$$

(a)

(b)

Fig. 14. Temperature and confining pressure versus time in laboratory test [28] and DEM simulation (a)

thermal recovery; (b) depressurization

To reproduce the stress conditions in laboratory, a servo stress-controlled boundary system was used in these DEM simulations. In the thermal recovery simulation, MH bond parameters were updated with the increase of temperature. In the depressurization simulation, change of back pressure was simulated by the change of effective confining pressure σ' as indicated in Fig. 14(b). In the meanwhile MH bond parameters were updated based on the changing back pressure, which was the confining pressure for MH bond.

It is noted that, when MH dissociates in nature, the temperature surrounding the dissociation zone decreases which delays the process of dissociation. However, in the current simulations, it is assumed that MH will dissociate as soon as the test condition reaches the stability boundary line, not taking the decrease of temperature into consideration. Once MH dissociates, methane hydrate cannot re-generate when the back pressure is restored as methane gas has escaped.

3.3 Macro-scale mechanical responses of thermal recovery tests

Fig. 15 shows the evolutions of deviator stress with axial strain in the DEM simulations and

laboratory tests [28] during thermal recovery. The initial dissociation points represent the time when the temperature started to increase. Fig. 16 shows the changes of axial strain with time in the DEM simulations and laboratory tests [28], while Fig. 17 shows the variations of volume change versus axial strain in the DEM simulations and laboratory tests [28].

Fig. 15. Stress-strain response in (a) DEM simulation and (b) laboratory test[28] during thermal recovery

It can be observed that in the DEM simulations, if the deviator stress applied is zero (sample *a*, only confining pressure applied), the axial strain increases to 1.5% with the increase of temperature accompanying MH dissociation, and then remains constant once the MH dissociates completed. If the deviator stress lies between zero and the strength of pure sand sample (sample *b*), the axial strain increases to 4% with increasing temperature and then remains constant on the completion of MH dissociation. If the deviator stress is larger than the peak deviator stress of pure sand sample (sample *c*), the axial strain increases rapidly with MH dissociation and the deviator stress cannot be maintained at the specified value as the sample has collapsed without the strengthening of MH bond. All the three samples experience a volumetric contraction but the void ratio was slightly larger than the pure sand sample at the same axial strain. By comparing all results, the mechanical responses of MH in the DEM simulations are found to be similar to those

in the laboratory tests, which implies that the proposed model can capture the features of dissociation caused by thermal recovery. Note that the deformation of MHBS is not only caused by the changes of effective stress but also by MH bond. With temperature increasing, the cage type skeleton of MH becomes unstable and in turn causes the decreases of MH bond strength and stiffness which will affect the behavior of MHBS.

(a)

(b)

Fig. 16. Changes of axial strain with time in (a) DEM simulation and (b) laboratory test [28] during thermal recovery

(a)

Fig. 17. Changes of volume with axial strain in (a) DEM simulation and (b) laboratory test [28]
during thermal recovery

3.4 Macro-scale mechanical responses of depressurization tests

Fig. 18 illustrates the relationship between deviator stress ratio q/p' (q – deviator stress; p' – mean effective stress) and axial strain in the DEM simulations and laboratory tests [28] during MH dissociation caused by depressurization. The initial dissociation points represent the time when the back pressure started to decrease, and a' , b' and c' represent the time when the back pressure were raised for sample a , b and c , respectively. Fig. 19 illustrates the evolutions of axial strain during depressurization and re-pressurization in the DEM simulations and laboratory tests [28], while Fig. 20 shows the relationship between volume change and axial strain in the DEM simulations and laboratory tests [28]. These figures show that (1) if the deviator stress is zero (sample a), the axial strain increases to 3.8% and the planar void ratio decreases from 0.24 to 0.14 with decreasing back pressure, while the deviator stress ratio q/p' mobilizes about zero; (2) if the deviator stress is between zero and the strength of pure sand sample (sample b), after depressurization, the axial strain increases to 5.6% and the planar void ratio decreases to 0.13 with constant deviator stress, while the deviator stress ratio q/p' decreases from 0.55 to 0.15 and then increases to 0.28 at b' ; (3) if the deviator stress is larger than the strength of pure sand sample (sample c), after depressurization, the axial strain increases to 8.2% and the planar void ratio decreases to 0.11 with constant deviator stress, while the deviator stress ratio q/p' decreases from 0.8 to 0.35 and then increases to 0.46 at c' .

After re-pressurization (all of these three samples were pure sand samples then), the applied effective confining pressure σ' returned back to its initial values. The macro mechanical properties of these three samples during the re-pressurization process have great differences: (1) If the initial deviator stress is zero (sample a), axial strain decreases quickly to a near zero value which is the same as the pure sand sample with no deviator stress; (2) if the deviator stress is between zero and

the strength of pure sand sample (sample *b*), the axial strain decreases and then remains constant, which is 4.5% in the DEM simulation and about 2.5% in the laboratory test, while the deviator stress ratio and planar void ratio increase until approach those of the pure sand sample; (3) if the deviator stress is larger than the compression strength of pure sand sample (sample *c*), the axial strain decreases slightly first, and then increases rapidly until sample *c* reaches the residual state. The deviator stress ratio increases first, and once it reaches a value that is larger than the peak deviator stress of pure sand, it begins to decrease and approaches to that of pure sand. The planar void ratio increases first, and once it reaches a value smaller than that of pure sand, it remains constant with increasing axial strain. The reason of previous phenomenon is that, there are two causes of plastic volumetric change: one is the increase of the effective mean stress, and the other one is shearing deformation as illustrated by the well known stress-dilatancy phenomenon, both of which come from the rearrangement of particles instead of particle deformation. After re-pressurization, methane hydrate bonds have vanished and methane hydrate bearing sediment sample becomes a pure sand sample. A pure sand sample is weaker than the methane hydrate bearing sediment sample. Although the mean stress and the stress ratio have gone back to its initial value, the deformation increases obviously accompanied by plastic shear deformation and shear-induced volumetric strain, if the deviator stress is above the peak deviator stress of a pure sand.

By comparing these results, the mechanical responses in the DEM simulations are found to be similar to those in the laboratory tests, which validated the proposed MH bond model on simulating the process of dissociation caused by depressurization.

Fig. 18. Stress-strain response in (a) DEM simulation and (b) laboratory test[28] during MH dissociation caused by depressurization

Comparing Fig. 19(b) with Fig. 20(b), the changes of axial strain with time are different although they were derived from the same laboratory test [28]. After re-pressurization, the axial strain of sample *b* has a decrease tendency in Fig. 19(b) while an increase tendency in Figs. 18(b) and 19(b). The authors believe that the measurement of axial strain should be examined further. The evolution of axial strain shown in Fig. 19(b) [28] is consistent with the DEM simulation results. In actual undersea condition, the back pressure (pore water pressure) and the total confining pressure changes equally, i.e., the effective stress is constant with sea level changing. With sea level increasing, the back pressure (pore water pressure) increases and the strength and stiffness of MH bond increase with no obvious deformation of MHBS being caused. With sea level decreasing, the strength and stiffness of MH bond decrease until vanish as the cage type skeleton of methane hydrate becomes unstable, and deformation of MHBS may occur in this condition.

Fig. 19. Changes of axial strain with time in (a) DEM simulation and (b) laboratory test [28] during MH dissociation caused by depressurization

Fig. 20. Changes of volume with axial strain in (a) DEM simulation and (b) laboratory test [28] during MH dissociation caused by depressurization

4 Further observations of micro-scale mechanical responses of thermal recovery tests

Micro-scale parameters, which can be analyzed by PFC2D, are as important as macro-scale mechanical responses in geotechnical research. In this paper, three micro-scale parameters, contact distribution, APR, and MH bond ratio distribution, were analyzed as follows.

4.1 Contact distribution

Contact distribution demonstrates the spatial arrangement of contacts, stress transmission paths and changes of soil skeleton. It has important effects on the macro-scale mechanical responses. Dividing a circle into 36 sectors with 10 degrees each, the ratio of the number of contacts in each sector to the total number of contacts could be determined. Then the contact distribution diagram could be drawn.

There are two types of contacts in MHBS, i.e., the bonded contact and the unbonded contact. In order to study the effects of temperature and applied deviator stress during thermal recovery, the distributions of the total, bonded and unbonded contacts in samples a , b and c at characteristic moment B ($t=0.5h$ in Fig. 14(a)) were analyzed and shown in Fig. 21. It can be seen that with increasing deviator stress, the total contact distribution changes from slight anisotropy to apparent

anisotropy. The portion of unbonded contacts increases with increasing deviator stress as the enclosed area of unbonded contact distribution keeps increasing. The distribution of unbonded contact remains anisotropic with a vertical major principal direction, which is consistent with the direction of major principal stress since more particle contacts are required to resist the higher vertical stress. The portion of bonded contacts decreases and its distribution remains anisotropic with a horizontal major principal direction, which is perpendicular to the major principal direction of unbonded contact distribution. For sample *a* with zero deviator stress (i.e., only confining pressure applied), very few bonds were damaged despite the increasing of temperature for 0.5 hours (Fig. 21(a)). The contact distribution was similar to the initial condition ($t=0h$), when the bonds were formed at $\sigma'=0.2MPa$. It is noted that the initial total contacts in sample *a* shows slight anisotropy (similar to Fig. 21(a)) caused by specimen compaction. This may reflect the actual anisotropic condition of nature MHBS.

However for sample *b* with same bond strength as sample *a*, MH bonds were damaged apparently. This difference was caused by the application of deviator stress. As the deviator stress of 3.8MPa was applied, forces transmitted in sample *b* were larger than in sample *a*, thus many vertical bonded contact were damaged and bonded contact distribution showed anisotropy with a horizontal major principal direction. By comparing sample *b* and sample *c*, it is seen that larger force causes more MH bond damages as the area of bonded contact distribution decreases, and the anisotropy of MHBS is more evident.

The total, bond and unbonded contact distributions of sample *c* at three characteristic moments *A* ($t=0h$, initial condition), *B* ($t=0.5h$, during dissociation) and *C* ($t=2h$, after dissociation) were also analyzed and shown in Fig. 22. It can be seen that with increasing temperature, the degree of anisotropy of the total contacts increases with its major principal direction remains in vertical. The portion of bonded contacts decreases with a horizontal major principal direction, while the portion of unbonded contacts increases with a vertical major principal direction. When the MH bonds dissociate completely ($t=2h$), bonded contacts disappear and unbonded contacts are the same as the total contacts.

Sample <i>a</i>	Sample <i>b</i>	Sample <i>c</i>
(a)	(b)	(c)

Fig. 21. Contact distribution of different samples at $t=0.5h$ during thermal recovery

At $t=2h$ and $T=293K$ (point *C* in Fig. 14(a)), the test condition is in the unstable zone, thus MH bonds has dissociated completely. Sample *c* has collapsed without the strengthening of MH bond as the deviator stress applied is larger than the compression strength of pure sand sample. In order to resist the specified deviator stress, vertical inter particle contacts increase more than horizontal ones. Thus degree of anisotropy is higher than that shown in Fig. 22(b). At $t=2h$, more particle movements and rotations (see APR analyses below) occurred which caused the shrunk of large void and soil contraction as shown in Fig. 17(a).

4.2 APR and bond ratio distribution

On consideration of energy dissipation, Jiang et al. [41] decomposed general contact displacement into two components: rolling and sliding, which have relationships with particle translational movement, rotation and particle size. The effects of particle rotation and particle size can be illustrated by the averaged pure rotation rate (APR), which can be expressed as

$$\omega = \frac{1}{N} \sum_{k=1}^N \left[\frac{1}{\bar{R}} (\theta_1^k R_1^k + \theta_2^k R_2^k) \right] \quad (38)$$

where ω is the averaged pure rotation rate; N is the total contact number in the representative volume; the superscript k represents the k^{th} contact; $\bar{R}$ is the same as that in Eq (3); θ_1^k, θ_2^k are rotational velocities of two contacting particles. The value of APR can be used to indicate not only the energy dissipation, but also the variation of micro structures of soil.

In order to show the effect of MH bonds, the bond ratio was also analyzed to investigate the micro structure change. The bond ratio R_b can be expressed as

$$R_b = \frac{N_b}{N_c} \quad (39)$$

where N_c is the initial number of MH bond and N_b is the remain bond number.

(a) A ($t=0$ h) (b) B ($t=0.5$ h) (c) C ($t=2$ h)

Fig. 22. Contact distribution of Sample c at different time

Fig. 23 depicts the APR distributions of sample a , b and c at $t=0.5$ h during thermal recovery. It shows that with increasing deviator stress, the maximum absolute value of APR increases, and the distribution of positive and negative values of APR become increasingly localized, which indicates shear bands developed there. Fig. 24 depicts the bond ratio distribution of sample a , b and c at $t=0.5$ h. It shows that the value of bond ratio decreases with increasing deviator stress, which implies that there are more bonds damaged under higher deviator stress, which is consistent with the observation of the area of bonded contact distribution in Fig. 21.

For sample a , the axial strain is 0.05% at $t=0.5$ h as shown in Fig. 16(a), and only a few bonds are damaged. Many particles are still bonded by MH bond and are difficult to rotate, which causes the APR values rather small. For sample b , the axial strain is 2.6% at $t=0.5$ h as shown in Fig. 16(a), some MH bonds were damaged on the application of deviator stress (3.8MPa), and some particles can rotate without the constrain of MH bonds, thus the APR values are higher, and sample b was compressed further than sample a . For sample c , the axial strain is 6.2% at $t=0.5$ h. At this time, more bonds were damaged compared with sample b on the application of higher deviator stress (6.8MPa). More particles can rotate, and the APR values increase further, and sample c is compressed further with several developed shear bands as consistent with the localized distribution of positive and negative values of APR.

Fig. 23. APR distributions of Samples a , b and c at $t=0.5$ h during thermal recovery

Sample *a*

Sample *b*

Sample *c*

Fig. 24. Bond ratio distribution of Samples *a*, *b* and *c* at $t=0.5h$ during thermal recovery

Fig. 25 shows the APR distributions of sample *c* at points *A* ($t=0.5h$), *B* ($t=0.5h$) and *C* ($t=0.5h$), corresponding to the initial condition, during thermal recovery (still in the stable zone) and after thermal recovery (MH dissociates completely), respectively, while Fig. 26 shows the distribution of the bond ratio in sample *c* at points *A*, *B* and *C*. Fig. 25 shows that with increasing temperature, the maximum absolute value of APR increases, and the distribution of positive and negative values of APR becomes increasingly localized, which indicates the development of shear bands. Fig. 26 shows that the value of bond ratio decreases with increasing temperature. At $t=0h$, sample *c* is still under the initial test condition, and MH bond strength and elastic modulus were not changed at this time. At $t=2h$, $T=293K$ and $\sigma'=5MPa$, the condition point is in the lower right zone to the stability boundary line, and MH dissociates completely and only pure sand left as shown in Fig. 26(c). As the applied deviator stress (6.8MPa) is larger than the compression strength, sample *c* has been damaged and reached the critical state at $t=2h$.

A ($t=0$ h)B ($t=0.5$ h)C ($t=2$ h)Fig. 25. APR distributions of Samples c at different time during thermal recovery Fig. 26. Bond ratio distribution of Sample c at different time during thermal recovery

5 Further observations in micro-scale mechanical responses of depressurization tests

5.1 Contact distribution

In order to study the micro-scale parameters during depressurization and re-pressurization, the total contact distribution of sample a , b and c at point $B(t=4h)$ were analyzed. Moreover, the total contact distributions at points $A(t=0h)$, $B(t=4h)$ and $C(t=7h)$ of sample c were also studied, corresponding to the initial condition, after depressurization and after repressurization, respectively. As the back pressure was reduced very quickly, no analysis was conducted during the process of depressurization. Moreover, as the MH bond has dissociated completely at B and C , there is no analysis on MH bond distribution herein.

Fig. 27 illustrates the total contact distributions of sample a , b and c at $t=4h$ after depressurization. It shows that the total contact distributions of samples under different deviator stress are nearly the same which is different from the effect of deviator stress during thermal recovery as shown in Fig. 21. The reason is that samples during thermal recovery are still partially bonded by MH bond, and higher deviator stress causes more bond breakage. However samples at $t=4h$ are pure sand samples ($T=293K$ and $\sigma_w=3.5MPa$, at the lower right zone of the stability boundary line), so their contact distribution are not affected by MH bond breakage. Note that,

although the deviator stress applied is larger than the compression strength of pure sand, sample *c* was not damaged as the mean effective stress increased with decreasing back pressure.

Fig. 28 illustrates the total contact distributions at $t=0\text{h}$, $t=4\text{h}$, and $t=7\text{h}$ of sample *c* during depressurization process. It shows that at $t=0\text{h}$ and $t=4\text{h}$, the total contact distributions show slight anisotropy, while the anisotropy distribution is more apparent at $t=7\text{h}$. The total contact distribution at $t=0\text{h}$ of sample *c* is the same as the sample *c* at $t=0\text{h}$ in thermal recovery. At $t=4\text{h}$, the effective confining pressure is increased to 11.5MPa as the back pressure is decreased, and sample *c* is compressed further, thus the degree of anisotropy of the contact distribution shows a slight increase. At $t=7\text{h}$, the effective confining pressure is reduced back to 5MPa and sample *c* has been damaged. Soil particle rearrangements become more rigorous as shown in the following APR analysis, and there are more vertical contacts to resist the higher deviator stress ratio applied.

Fig. 27. Total contact distributions of Samples *a*, *b* and *c* at $t=4\text{h}$ during depressurization

Fig. 28. Total contact distributions of Sample *c* at different time during depressurization

5.2 APR distribution

Fig. 29 illustrates the APR distributions of sample *a*, *b* and *c* at $t=4\text{h}$ after depressurization. It shows that with increasing deviator stress, the maximum absolute value of APR increases. These three samples are pure sand samples at $t=4\text{h}$, and the difference is the deviator stress applied. The axial strain of sample *a*, *b* and *c* are 3.3%, 5% and 8.6%, respectively. Samples are compressed further with

higher deviator stress, and the rearrangements of particles such as particle rotations become more rigorous.

Fig. 30 illustrates APR distribution of sample *c* at $t=0$, $t=4$ h and $t=7$ h. It shows that the maximum absolute value of APR increases with back pressure being decreased from 10MPa to 3.5MPa (effective confining pressure being increased from 5MPa to 11.5MPa) during 0h~4h. Sample *c* is compressed further as mean effective stress increases. It is also shown that with back pressure being increased back to 10MPa (4h~7h), the maximum absolute value of APR increases further and the positive and negative values of APR become increasingly localized indicating the developments of several shear bands at $t=7$ h. At $t=7$ h after repressurization, stress condition returns to its initial values, and sample *c* approaches critical state at this moment. The soil skeleton becomes unstable, and the rearrangement of particles becomes rigorous, therefore the APR values are larger than before.

Fig. 29. APR distributions of Samples *a*, *b* and *c* at $t=4$ h during depressurization

Fig. 30. APR distribution of Sample *c* at different time during depressurization

6 Conclusions

A new thermo-hydro-mechanical bonded contact model was proposed and then implemented into PFC2D to simulate the MHBS exploitation by thermal recovery and depressurization methods. The macro-scale responses were compared with the laboratory tests conducted by Hyodo et al [28] to validate the new model, and the micro-scale parameters were also analyzed. It has been found that:

(1) The macro-scale mechanical responses of three samples in DEM simulations and laboratory tests during thermal recovery exhibit similar characteristics, such as volume contraction, strain softening, which implies that the newly proposed model is applicable to qualitatively capture the features of thermal dissociation process of MHBS, although the accurate design values cannot be determined using PFC2D.

(2) Similarly, the macro-scale mechanical responses of three samples during depressurization and repressurization process in the laboratory tests, including evolution of deviator stress ratio and planar void ratio, were also captured qualitatively in DEM simulations.

(3) MHBS sample will be damaged after exploitation (no matter thermal recovery or depressurization) when the deviator stress applied is larger than the compression strength of pure host sand.

(4) The changes of axial strain after repressurization as mentioned in [28] have some contradiction, i.e. the axial strain increases in Fig. 18(b) while decreases or remains constant in Fig. 19(b). The decrease of axial strain in DEM simulations matches that as observed in Fig. 19(b). This could be a point to be studied further.

(5) During the thermal recovery, the bond contact distribution changes apparently with increasing temperature or deviator stress with a horizontal major principal direction. MH bond contacts dissociate completely at the end of the process. The maximum absolute value of APR increases as the temperature or deviator stress increases.

(6) During the depressurization exploitation, the degree of anisotropy of contact distribution only changes slightly due to increase of deviator stress. The maximum absolute value of APR increases as the deviator stress increases.

(7) During the depressurization exploitation, although MH dissociates totally, sample with higher deviator stress than the compression strength is not damaged before re-pressurization and is

only damaged after the reinjection of pore water pressure. however, during the thermal recovery exploitation, MHBS is damaged as soon as MH dissociates. This indicates that the depressurization method is safer than the thermal recovery method.

In reality, the pore growth behavior of methane hydrate is also affected by salinity, which may somewhat alter the behavior of methane hydrate, which is still under research. Although the factors that influence the mechanical behavior of pure methane hydrate are not fully considered in this paper, the general behavior of methane hydrate can be reflected qualitatively.

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