Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2019.24202

168 (2019) 366–373
November

A practical method for predicting marine saturation of NGH

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Received 1 October 2018; Accepted 17 January 2019

ABSTRACT

Natural gas hydrate (NGH), a high-efficiency and clean unconventional energy, is an important alternative energy of fossil fuels in the future. 98% of the world's NGH is distributed in the sea area. BSR, anomalous attributes, and inversion techniques are widely used to predict the accumulation degree of NGH in the sea, but none of them can be used to directly predict NGH saturation. Therefore, we have relatively limited ability to represent the accumulation degree of NGH in the sea area. Although there are currently some methods for directly predicting the saturation of NGH, the method is based on test and no prediction result is obtained. The disadvantages of this method are the long calculation time and that the system error is not taken into account, and therefore it may lead to difficulty in practical promotion of the method. In view of the problems in the prediction of NGH saturation, a new idea of prediction has been developed in this research, with the main advantage that it can better eliminate the system error, thus to improve the prediction accuracy; this idea is then introduced to the existing three classic NGH saturation prediction methods; finally, based on the data of the Shenhu sea area of China, Wood method with the smallest prediction error is selected from the three prediction methods, which is the most applicable NGH saturation prediction method.

Keywords: NGH; Fossil energy; NGH saturation prediction method; System error; Wood method

1. Introduction

Natural gas hydrate (NGH) is an ice-like crystal compound formed by water and gas molecules under low temperature and high-pressure conditions. The gas molecule (object) is confined in the hydrogen bond cage formed by water molecules (subject), and the two are stably combined by Van der Waals force [1]. According to the prediction by scientists, the carbon content of energy existing in the form of NGH worldwide is approximately twice the total carbon content of the current proven fossil fuels [2]. The products of NGH combustion are carbon dioxide and water, and therefore NGH is a clean unconventional energy. It is an important alternative energy of fossil fuels in the future [3]. 98% of NGH is distributed in sea areas and only 2% in land areas worldwide. Therefore, the research on NGH in sea areas has attracted more concern and attention in the world. Major coastal countries around the world, such as the United States, Russia, China, India, Japan, Korea, etc., have conducted drilling coring of marine NGH and successfully obtained NGH samples. However, successful coring does not mean to find good NGH enrichment regions. Scholars have carried out some research work on NGH enrichment. For example, BSR, abnormal properties, inversion method,

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etc. have been used to find regions favorable for NGH enrichment [4–8]. The currently existing enrichment prediction methods are mainly qualitative methods, but there are also some quantitative prediction methods, for example, judging the NGH enrichment degree by using the relative wave impedance. But this method is essentially a qualitative method because it is unable to obtain the NGH saturation value from the results of this method, and therefore it is difficult to really find the absolute NGH enrichment degree. Quantitative NGH saturation prediction method is more conducive to find the enrichment degree of NGH. At present, NGH saturation calculation methods include time-average equation method, Timur time-average equation method, Wood correction equation method, Gassmann method, etc. [9–12]. Although the NGH saturation prediction methods have been developed early, their practical application is very rare. Guo et al. have carried out some applications in the Shenhu sea area of China to contrast the velocity of NGH-containing sediments and the measured velocity on the basis of assumed NGH saturation, and thereby judging the NGH saturation at different depths [13]. The disadvantages of this method include the low efficiency and difficulty in promotion. One of the major reasons why there are rare quantitative NGH saturation prediction methods at present is that the actual situation is more complex than the assumptions of the method used, resulting in a great difference between predicted and measured results. Analyzing the reason for the difference between predicted and measured result and developing a solution is the prerequisite and basis for the successful application of the current NGH saturation rock theory. According to the analysis of the actual data of the Shenhu sea area in South China Sea, there are two major reasons for the great difference between the two: (1) it is very difficult to measure the actual parameters, such as material composition and proportion of skeleton, porosity, density of sediment, etc.; (2) the calculation of elastic parameters with the methods is based on certain assumptions, which have certain deviations compared with the actual situation. Error caused by the first reason can be regarded as a system error which can be eliminated by system correction, while the error caused by the second reason is a method error which cannot be eliminated directly. Given this understanding, this paper conducts research on the selection of NGH saturation prediction method from the three classic methods on the basis of the data of Shenhu sea area in the South China Sea, so as to promote the development of NGH saturation prediction method for the South China Sea area, specifically as follows.

2. NGH saturation prediction method

2.1. Three classic NGH saturation prediction methods

Influenced by gas sources, temperature and pressure, and geology, the saturation of marine NGH is often less than 100% in most cases. Therefore, it is more practical to choose suitable prediction methods for different NGH saturations. At present, there are three major methods to predict different NGH saturations. (1) Timur time-average equation method: In 1968, Timur proposed a three-component time-average model to explain the longitudinal wave velocity in rocks with different degrees of cementation at permafrost temperatures [14]. This model is called Timur time-average equation and was first used by Person et al. in the study of different NGH saturations in 1983 [15]. (2) Wood correction equation method: it is a method proposed by Lee et al. for the study of different NGH saturations based on the study with Timur time-average equation method. (3) Gassmann equation method for marine NGH-containing sediments: It is a method proposed by Helgerud et al. in 1999 for the study of different NGH saturations [16].

2.1.1. Timur time-average equation

This method considers that the *P*-wave velocity of NGH-containing sediments should be related to porosity, NGH saturation, *P*-wave velocity of water in pores, *P*-wave velocity of NGH and *P*-wave velocity of rock skeleton. The relationship can be expressed by Eq. (1).

$$\frac{1}{\upsilon_b}=\frac{\phi(1-S_h)}{\upsilon_{\mathrm{pw}}}+\frac{\phi S_h}{\upsilon_{\mathrm{ph}}}+\frac{1-\phi}{\upsilon_{\mathrm{pm}}} \tag{1}$$

wherein υ_b – the *P*-wave velocity of NGH-containing sediments; ϕ – porosity; S_h – the saturation of NGH; υ_{pw} – the *P*-wave velocity of water in pores; υ_{ph} – the *P*-wave velocity of pure NGH; υ_{pm} – the *P*-wave velocity of rock skeleton.

2.1.2. Wood correction equation

This method considers that, in addition to porosity, NGH saturation, *P*-wave velocity of seawater, *P*-wave velocity of NGH and *P*-wave velocity of rock skeleton, the *P*-wave velocity of NGH-containing sediments should also be related to the density of NGH-containing sediments, the density of water in pores, the density of NGH and the density of rock skeleton. The relationship can be expressed by Eq. (2).

$$\frac{1}{\rho_b\upsilon_b^2}=\frac{\phi(1-S_h)}{\rho_w\upsilon_{\mathrm{pw}}^2}+\frac{\phi S_h}{\rho_h\upsilon_{\mathrm{ph}}^2}+\frac{1-\phi}{\rho_m\upsilon_{\mathrm{pm}}^2} \tag{2}$$

wherein ρ_b – formation density; ρ_w – the density of water in pores; ρ_h – the density of NGH; ρ_m – the density of rock skeleton.

2.1.3. Gassmann equation for marine NGH-containing sediments

High-porosity marine sediments can be considered as a "granular system", and its *P*-wave velocity has the following relationship with the sediment bulk modulus, shear modulus and density of saturated fluid [12,17]:

$$\upsilon_b=\sqrt{\frac{K_{\mathrm{sat}}+\frac{4}{3}G_{\mathrm{sat}}}{\rho_b}} \tag{3}$$

wherein K_{sat} – the sediment bulk modulus of saturated fluid; G_{sat} – sediment shear modulus.

According to Gassmann equation, the relationship between the sediment bulk modulus, shear modulus of saturated fluid and the bulk modulus, shear modulus and porosity of dry rock, the bulk modulus of fluid and the bulk modulus of rock skeleton can be expressed by Eqs. (4) and (5) [12,16].

$$K_{\text{sat}} = K\frac{\phi K_d - (1+\phi)K_f K_d / K + K_f}{(1-\phi)K_f + \phi K - K_f K_{d/K}} \tag{4}$$

$$G_{\text{sat}} = G_d \tag{5}$$

wherein K – the bulk modulus of rock skeleton; K_d – the bulk modulus of dry rock; K_f – the bulk modulus of fluid; G_d – shear modulus of dry rock.

The bulk modulus and shear modulus of rock skeleton can be found with Hill (1952) Average equation [16].

$$K = \frac{1}{2}\left[\sum_{i=1}^{m} f_i K_i + \left(\sum_{i=1}^{m}\frac{f_i}{K_i}\right)^{-1}\right] \tag{6}$$

$$G = \frac{1}{2}\left[\sum_{i=1}^{m} f_i G_i + \left(\sum_{i=1}^{m}\frac{f_i}{G_i}\right)^{-1}\right] \tag{7}$$

wherein f_i is the volume percentage of the ith mineral constituent in rock skeleton; K_i is the bulk modulus of the ith mineral constituent; G is the shear modulus of dry rock; G_i is the shear modulus of the ith mineral constituent.

The bulk modulus of fluid can be found with the Reuss (1929) average equation [16].

$$K_f = \frac{S_w}{K_w} + \frac{1-S_w}{K_h} \tag{8}$$

For marine sediments with high porosity, the bulk modulus and shear modulus of dry rock can be calculated under two conditions. One is that the porosity is smaller than the critical one, and the other is that the porosity is greater than the critical one.

When the porosity is smaller than the critical one, the bulk modulus and shear modulus of dry rock can be calculated with Eqs. (9) and (10).

$$K_d = \left[\frac{\phi/\phi_c}{K_{\text{HM}} + \frac{4}{3}G_{\text{HM}}} + \frac{1-\phi/\phi_c}{K + \frac{4}{3}G_{\text{HM}}}\right]^{-1} - \frac{4}{3}G_{\text{HM}} \tag{9}$$

$$G_d = \left[\frac{\phi/\phi_c}{G_{\text{HM}} + Z} + \frac{1-\phi/\phi_c}{G + Z}\right]^{-1} - Z \tag{10}$$

When the porosity is greater than the critical one, the bulk modulus and shear modulus of dry rock can be calculated with Eqs. (11) and (12).

$$K_d = \left[\frac{(1-\phi)/(1-\phi_c)}{K_{\text{HM}} + \frac{4}{3}G_{\text{HM}}} + \frac{(\phi-\phi_c)/(1-\phi_c)}{\frac{4}{3}G_{\text{HM}}}\right]^{-1} - \frac{4}{3}G_{\text{HM}} \tag{11}$$

$$G_d = \left[\frac{(1-\phi)/(1-\phi_c)}{G_{\text{HM}} + Z} + \frac{(\phi-\phi_c)/(1-\phi_c)}{GZ}\right]^{-1} - Z \tag{12}$$

K_{HM}, G_{HM} and Z in Eqs. (9)–(12) can be calculated with Eqs. (13)–(15).

$$K_{\text{HM}} = \left[\frac{G^2 n^2 (1-\phi_c)^2}{18\pi^2 (1-\upsilon)^2}\right]^{\frac{1}{3}} \tag{13}$$

$$G_{\text{HM}} = \frac{5-4\upsilon}{5(2-\upsilon)}\left[\frac{3G^2 n^2 (1-\phi_c)^2}{2\pi^2 (1-\phi)^2}P\right]^{\frac{1}{3}} \tag{14}$$

$$Z = \frac{G_{\text{HM}}}{6}\left[\frac{9K_{\text{HM}} + 8G_{\text{HM}}}{K_{\text{HM}} + 2G_{\text{HM}}}\right] \tag{15}$$

wherein n is the number of particles each particle contacts, which is generally taken as 8.5; P is effective pressure and can be calculated with Eq. (16).

$$P = (\rho_b - \rho_f)gD \tag{16}$$

wherein ρ_b – the density of sediments; ρ_f – the density of water; g – gravity acceleration; D – the depth of submarine sediments.

2.2. Steps of NGH saturation calculation

In this paper, the following steps of NGH saturation calculation are adopted to overcome the systematic errors of various methods, specifically as follows:

- Basic data preparation. These data include: (1) the relevant elastic parameters, such as mineral composition of rock skeleton, the bulk modulus and shear modulus of minerals, the density of minerals, the bulk modulus and density of seawater, the bulk modulus, shear modulus and density of pure NGH, etc.; (2) the relevant borehole data (the data must be continuous and include NGH zone and non-NGH zone above), such as the depth, P-wave velocity, porosity and density of sediments;
- The calculation of pre-correction saturation. Select a method to calculate NGH saturation. Suppose the saturation of NGH at a certain depth is S_{hi}^1, and i denotes depth sampling number.
- The calculation of NGH saturation correction amount. Calculate the average of S_{hi}^1 from non-NGH zone calculation. Suppose the average value is S_h^A, and then subtract 0 from S_h^A to obtain their difference; suppose the difference is ΔS_h.
- The calculation of final NGH saturation. Suppose the final NGH saturation is denoted by S_{hi}^2, then $S_{\text{hi}}^2 = S_{\text{hi}}^1 - \Delta S_h$.

For the convenience of description, the method of calculating the final NGH saturation based on Timur time-average equation is called Timur method; the method of calculating the final NGH saturation based on Wood correction equation is called Wood method; and the method based on Gassmann equation for marine NGH-containing sediments is called Gassmann method.

3. Application effect of the three methods in the Shenhu Sea area of China

3.1. Geological conditions of the research area

The research area is located in the Shenhu sea area at the northern margin of the South China Sea (the red area in Fig. 1 and belongs to Zhuer Depression of Pearl River Estuary Basin according to tectonic division [11]. From April to June 2007, China Geological Survey successively performed NGH drilling in places with BSR mark in the area (the position marked with a red circle in Fig. 2, and obtained NGH samples at some drilling locations, such as SH-2, SH-3 and SH-7, while no NGH sample was obtained at some other locations, such as SH-1 and SH-4 [5]. The three boreholes where NGH samples were obtained are located at the ridge of rugged sea bottom at the continental slope about 1,200 m deep. The NGH samples were roughly distributed in the pores of sediments approximately 200 m below the sea bottom. The components of sediments near the near samples are divided into three kinds according to the particle size [18,19]: (1) sand, particles with the particle size greater than 0.063 mm; (2) silt, particles with the particle size between 0.04 and 0.063 mm; (3) clay, particles with the particle size smaller than 0.04 mm. The major component of sediments is silt (with an average content of approximately 75%), followed by clay (20%) and sand (5%).

3.2. Calculation of seabed sediment porosity

In 2011, Guo et al. conducted laboratory analysis on the porosity of drilling samples from the Shenhu sea area and found that the seabed sediment porosity and depth have the approximate relationship as shown in Eq. (17) [13].

$$\phi = -7.44 \times \ln D + 85.38 \tag{17}$$

wherein ϕ is porosity, %; D is the depth of sediments on the seabed, in *m*.

3.3. Logging curve of S borehole

Figs. 3 and 4 show the *P*-wave velocity and density curves of one of the boreholes (suppose it is *S* borehole) where NGH was found by drilling. According to the acoustic logging curve of the *S* borehole, when the depth is between 200 and 220 m, the *P*-wave velocity increased significantly, up to 2,300 m/s, while the density had an apparent decrease. This location has been identified to have NGH [21].

3.4. Estimated NGH saturation based on chloride ion

The formation and decomposition of NGH will lead to change of the chloride ion concentration. When NGH is formed, it will reject salt ions dissolved in the water in pores and make the ions spread out; when NGH is decomposed, water in pores will be diluted, making the NGH samples obtained by drilling have low salinity than samples containing no NGH. NGH with different saturations tends to have different concentrations of chloride ions, so NGH saturation can be calculated based on the chloride ion concentration anomaly in pores. Fig. 5 shows *S* borehole NGH saturations obtained based on the chloride ion concentration anomaly [21]. The contrast between Figs. 3 and 5 shows that NGH saturation is relatively higher at the depth with higher *P*-wave velocity, which can be more than 30% and up to 50%.

3.5. Calculation of skeleton elastic parameters

With reference to the aforesaid analysis of the particle types of NGH-containing sediments, suppose the particle composition of sediment skeleton is as follows: siltstone 75%, clay 20% and sandstone 5%. Table 1 shows the bulk modulus,

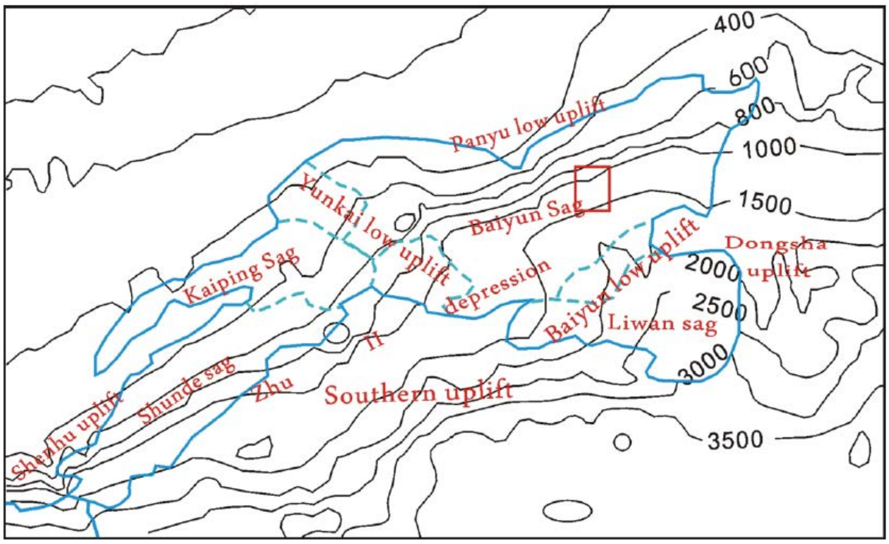

Fig. 1. Structural location of the research area.

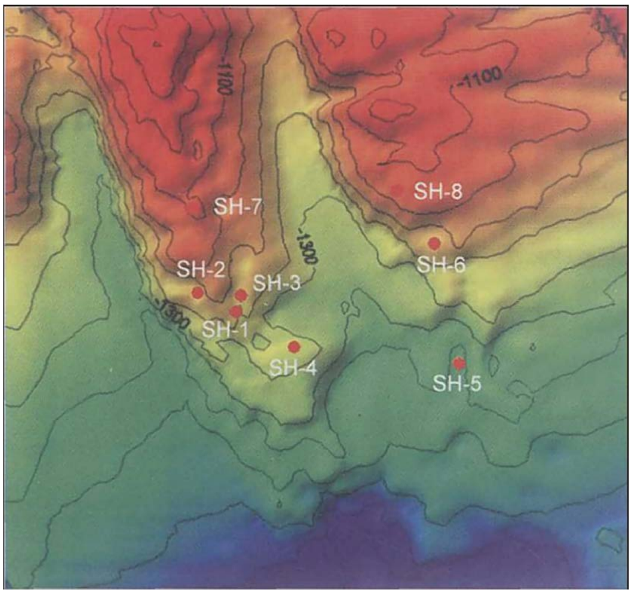

Fig. 2. Topography of the hydrate sampling area in the Shenhu sea area of the northern South China Sea [20].

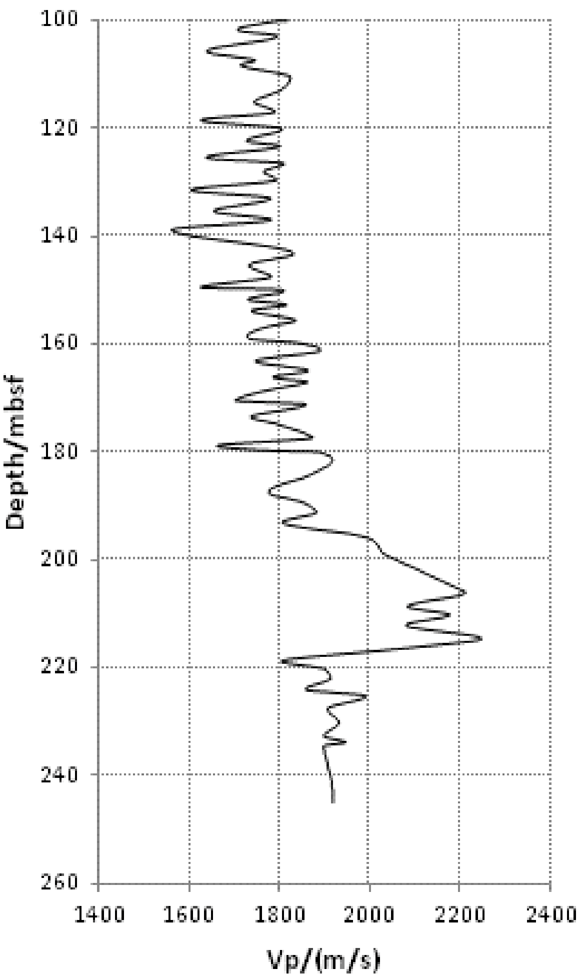

Fig. 3. *P*-wave velocity-depth curve of *S* borehole.

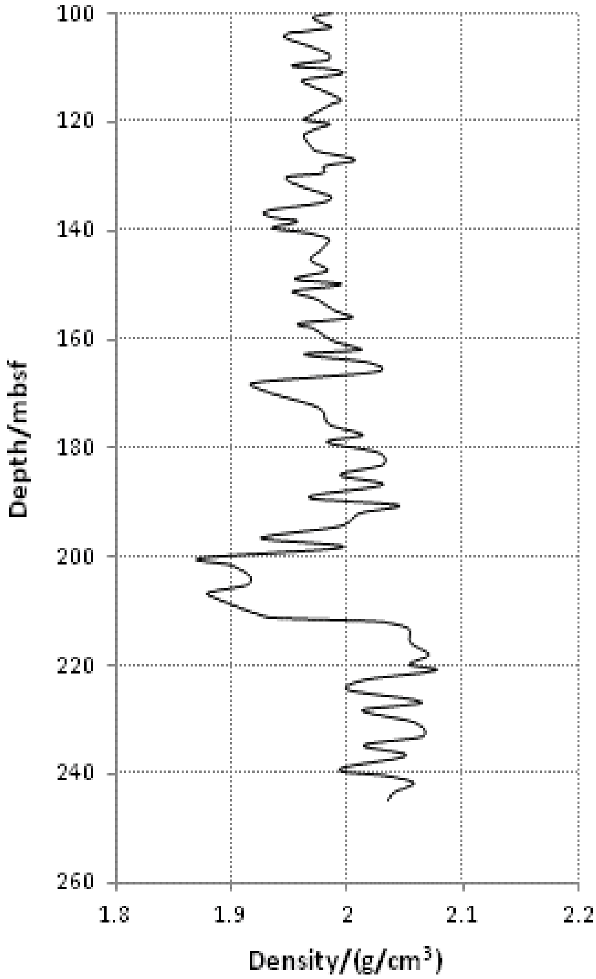

Fig. 4. Density-depth curve of *S* borehole.

shear modulus and density of siltstone, sandstone, clay, seawater and pure methane hydrate [20–22].

The components of sediment skeleton have three particle sizes. According to Eqs. (6) and (7), the bulk modulus and shear modulus of the skeleton can be expressed by Eqs. (18) and (19).

$$K = \frac{1}{2}\left[\sum_{i=1}^{3} f_i K_i + \left(\sum_{i=1}^{3}\frac{f_i}{K_i}\right)^{-1}\right] \tag{18}$$

$$G = \frac{1}{2}\left[\sum_{i=1}^{3} f_i G_i + \left(\sum_{i=1}^{3}\frac{f_i}{G_i}\right)^{-1}\right] \tag{19}$$

wherein f_1, f_2 and f_3 are the volume percentage of siltstone, sandstone and clay, respectively, in the skeleton. K_1, K_2 and K_3 are the bulk modulus of siltstone, sandstone and clay, respectively. G_1, G_2 and G_3 are shear modulus of siltstone, sandstone and clay [23–25].

According to Eqs. (18) and (19) and Table 1, the bulk modulus and shear modulus of the skeleton can be calculated to be approximately 35 and 20.5 GPa.

The density of skeleton can be calculated with Eq. (20).

$$\rho_m = f_1\rho_1 + f_2\rho_2 + f_3\rho_3 \quad (20)$$

wherein f_1, f_2 and f_3 are the volume percentage of siltstone, sandstone and clay, respectively, in the skeleton. ρ_1, ρ_2 and ρ_3 are the density of siltstone, sandstone and clay, respectively.

According to Eq. (18) and Table 1, the density of skeleton can be calculation to be 2.64 g/cm^3.

3.6. Prediction results of the three methods

Figs. 6–8, respectively, show the contrast between NGH saturations predicted with Timur method, Wood method, Gassmann method and estimated with chloride ion concentration. As shown in Fig. 6, at the sediment depth smaller than 200 m, the NGH saturations predicted with Timur method are basically the same with those estimated with chloride ion method; at the sediment depth of 200—220 m, the NGH saturations predicted with Timur method are in a higher value zone and most values are higher than those estimated with chloride ion method; at the depth greater than 220 m, the NGH saturations predicted with Timur method are higher than those estimated with chloride ion method. As shown in Fig. 7, at the sediment depth smaller than 200 m, the NGH saturations predicted with Wood Method are basically the same with those estimated with chloride ion method; at the sediment depth of 200–220 m, the NGH saturations predicted with Wood method are in a higher value zone and most values are slightly smaller than those estimated with chloride ion method; at the depth greater than 220 m, the NGH saturations predicted with Wood method are higher than those estimated with chloride ion method [26–33]. As shown in Fig. 8, at the sediment depth smaller than 200 m, the NGH saturations predicted with Gassmann method are basically the same with those estimated with chloride ion method; at the sediment depth of 200–220 m, the NGH saturations predicted with Gassmann method are in a higher value zone and most values are higher than those estimated with chloride ion method, with an absolute value error greater than 20%; at the depth greater than 220 m, the NGH saturations predicted with Gassmann method are basically the same with those estimated with chloride ion method.

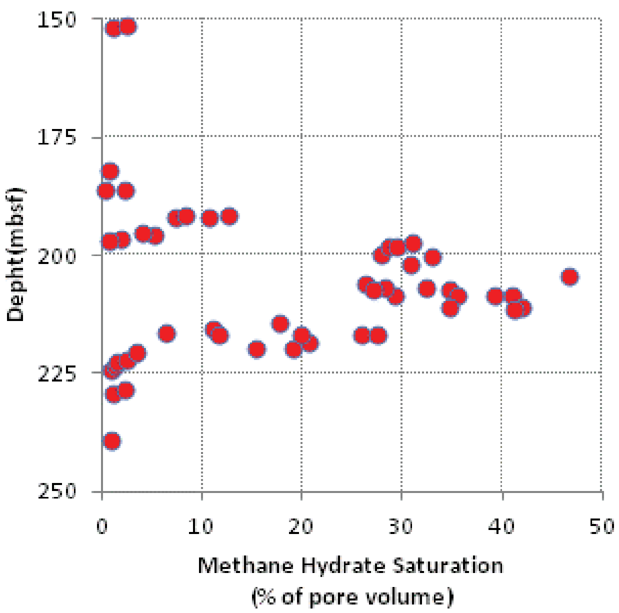

Fig. 5. *S* borehole NGH saturations estimated based on the chloride ion concentration anomaly [21].

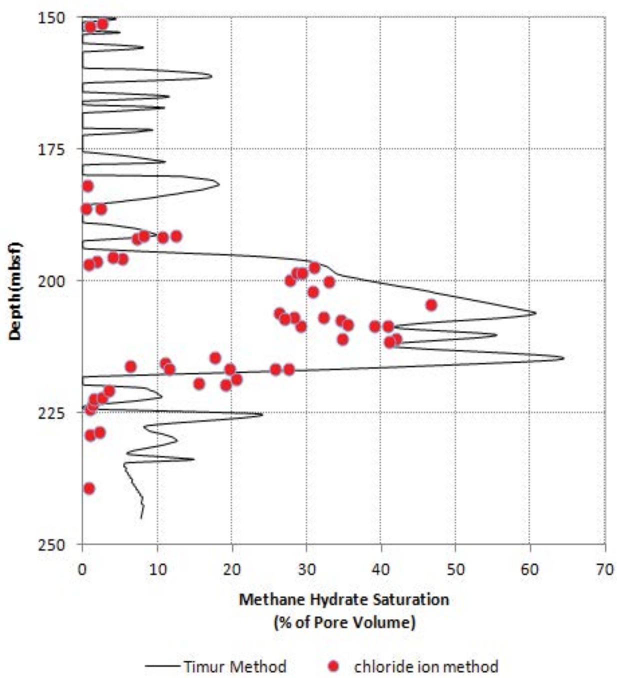

Fig. 6. Contrast between NGH saturations obtained with Timur method and chloride ion method.

Table 1
Elastic parameters of different components of sediments

Materials	Bulk modulus/GPa	Shear modulus/GPa	Density (g/cm^3)
Siltstone	40.08	28.13	2.65
Sandstone	40	25.06	2.65
Clay	20.9	6.85	2.58
Seawater	2.5	0	1.032
Pure methane hydrate	5.6	2.4	0.9

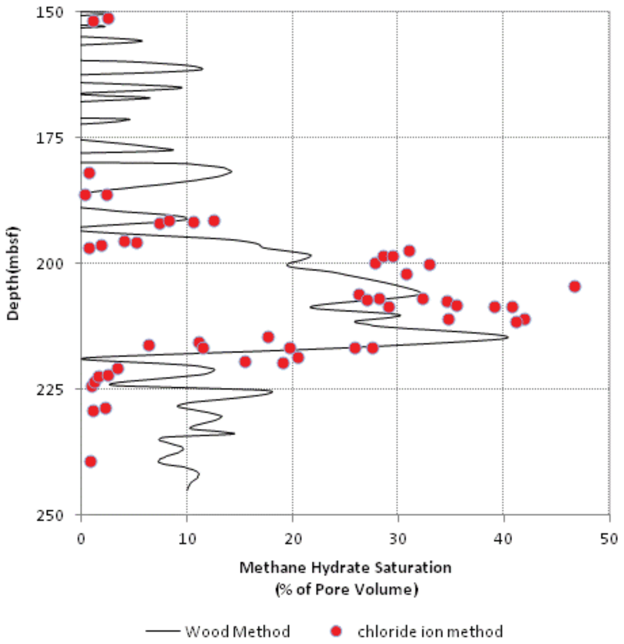

Fig. 7. Contrast between NGH saturations obtained with Wood method and chloride ion method.

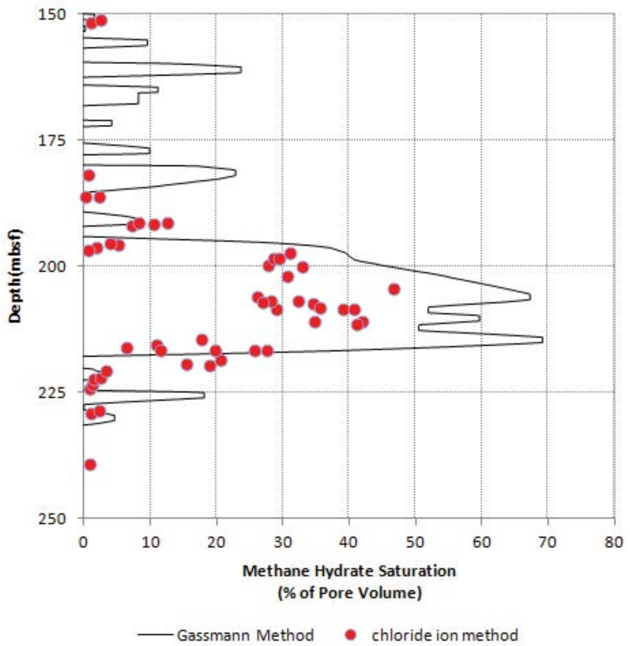

Fig. 8. Contrast between NGH saturations obtained with Gassmann method and chloride ion method.

4. Result analysis of the three methods

In this paper, comparisons are made based on the correlation coefficients and absolute value errors to select a relatively better method. Correlation coefficients are the prediction results of calculated between Timur method, Wood method, Gassmann method and chloride ion method to find the correlation coefficients of the three methods are 0.71, 0.64 and 0.74, respectively. Seen from the correlation coefficients, Gassmann method may be considered a good prediction method because it has the biggest correlation coefficient. In order to better evaluate the merits of the three methods, Fig. 9 shows the contrast of NGH errors of the three methods. It can be seen that most absolute value errors of Wood method are between –10% and 10%, and most value errors of Gassmann method are greater than 10%, indicating that Wood method is obviously superior to Gassmann method. The methods of evaluation with correlation coefficient and error bring different results. Which evaluation method is more reasonable? Research suggests that correlation coefficient is easily affected by sample size and typicality degree, while absolute value error has no relationship with the two factors. Therefore, absolute value error is taken as the reference standard for method selection.

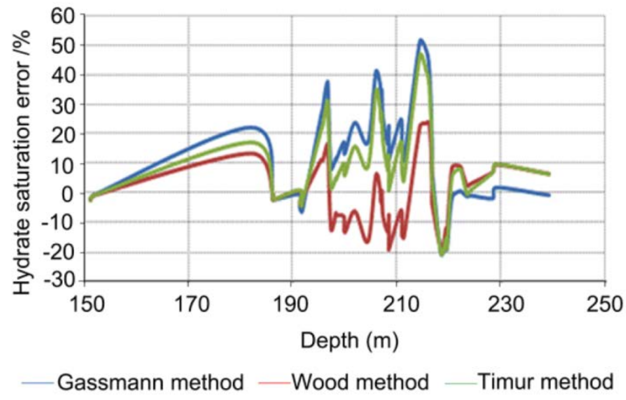

Fig. 9. Contrast between NGH errors of the three methods at different depths.

The average errors of Timur method, Wood method and Gassmann method are calculated to be 10.19, –0.35 and 13.32, respectively. The results show that Wood method has the smallest average error, so this method is a relatively good method for NGH saturation prediction.

5. Conclusions

Through the systematic research on the prediction methods of the saturation of NGH suspended in pores and contrast among different prediction methods, the following conclusions have been drawn:

- This paper has proposed an NGH saturation prediction method that can eliminate the errors introduced by the difference between actual measurement and theoretical assumption, to improve the effect of the prediction method in the actual application.
- This paper has proposed new NGH saturation calculations based on Timur time-average method, Wood correction equation and Gassmann equation for marine NGH-containing sediments, which are Timur method, Wood method and Gassmann method, respectively. The contrast among the three methods based on the actual data from the Shenhu sea area of China suggests that Wood method is a relatively good method for NGH saturation prediction.

- Wood method that can eliminate system errors is worth promotion for its smaller prediction error.

Acknowledgments

This research is supported by Open Fund (PLC20190604) of State Key Laboratory of Oiland Gas Reservoir Geology and Exploitation (Chengdu University of Technology), Open Fund of Guangdong Research Center for Unconventional Energy Engineering Technology (No. GF2018A001), and Guangdong "Sail Plan" Urgently Needed Personnel Introduction Program (Nos. 916022, 916023). This paper has been successfully completed, and we truly thanks Organization Department of Guangdong Province, Technology Department of Guangdong Province and Guangdong University of Petrochemical Technology for providing support to the first author (Xiangsheng Bao).

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