IMPROVED PREDICTION METHOD FOR GAS HYDRATE SATURATION IN SEA AREAS

Método mejorado de predicción para la saturación de hidrato de metano en áreas marinas

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ABSTRACT

Ninety-eight % of the world's natural gas hydrate (commonly known as hydrate) are distributed in the stability zone of the sea areas. There are also rich hydrates in the stability zone of the Shenhu Sea Area in China. Drilling in the Shenhu Sea Area of China reveals that a lot of hydrate exist in suspension mode. Now the Wood method is for predicting hydrate in suspension mode, but the practical application in the Shenhu Sea Area shows that this method leads to large errors in predicting hydrate saturation. In order to address the problem of low accuracy of the Wood method in predicting hydrate saturation in suspension mode, this paper has first pointed out that the low accuracy of the Wood method is primarily caused by the systematic analytic error of stratigraphic parameters, which is derived from insufficient understanding of sediment composition and porosity in sea area by existing analytic technology. The paper puts forward the Wood-SE method alternatively. The practical application in the Shenhu Sea Area shows that the absolute error of this method method is above 20%. The error of main hydrate occurrence section could even reach 60%.

Palabras clave: hidrato, zona de estabilidad,área marina de Shenhu, modo de suspensión, Método Wood, saturación de hidrato, método Wood-ES.

RESUMEN

El 98 % del hidrato de gas natural del mundo (comúnmente conocido como hidrato) se distribuye en la zona de estabilidad de las zonas marinas. También hay ricos hidratos en la zona de estabilidad de la zona del mar de Shenhu en China. La perforación en el área del mar de Shenhu de China revela que existe mucho hidrato en modo de suspensión. Ahora el método de madera es para predecir el hidrato en modo de suspensión, pero la aplicación práctica en el área del mar de Shenhu muestra que este método conduce a grandes errores en la predicción de la saturación de hidrato. Con el fin de abordar el problema de la baja precisión del método de la madera en la predicción de la saturación d

de hidrato en modo de suspensión, este documento ha señalado en primer lugar que la baja precisión del método de la madera es causada principalmente por el error analítico sistemático de los parámetros estratigráficos, que se deriva de una comprensión insuficiente de la composición de los sedimentos y la porosidad en la zona del mar por la tecnología analítica existente. El documento presenta el método Wood-SE alternativamente. La aplicación práctica en la zona del mar de Shenhu muestra que el error absoluto de este método se encuentra principalmente dentro del rango de -10%, 10%. La mayor parte del error absoluto del método Wood está por encima del 20%. El error de la sección principal de la ocurrencia del hidrato podría alcanzar incluso el 60 %.

INTRODUCTION

Natural gas hydrate (hydrate for short) is a kind of ice-like crystal compound formed by water and gas molecules under low temperature and high pressure, in which gas molecules (guest molecules) are confined in the hydrogen-bond cage formed by water molecules (host molecules), and the two exist stably through the combination of van der Waals force (Wu et al. 2017). According to the prediction by scientists, the carbon content of energy in the form of hydrate is about twice that of fossil fuels around the world (Fellet and Ramos 2015, Lu et al. 2019). The products of hydrate combustion are carbon dioxide and water. Therefore, hydrate is an efficient and clean unconventional energy and will be a vital alternative for fossil energy in the long run (Yang et al. 2020, Tang et al. 2016). Most of the hydrates are distributed in the sea areas, accounting for 98% of the total resources globally. The current studies on hydrate in the sea areas show that hydrate is not widely distributed in the stability zone, and the stability zone only determines the spatial scope of hydrate (Ruan et al. 2007, Liang et al. 2010, Luo, 2013, Yang et al. 2013, Zhang et al. 2014, Wan et al. 2016, Bao et al. 2018a). Hence, the presence of hydrate in one region does not mean that hydrate is enriched in the same region. In fact, numerous factors affect the enrichment of hydrate in a certain space of the stability zone, such as sufficient sources of gas, optimal conditions for gas migration and optimal conditions of reservoir. There are many methods for predicting gas hydrate enrichment (Ruan et al. 2007, Liang et al. 2010, Luo 2013, Yang et al. 2013, You et al. 2014, Zhang et al. 2014, Wan et al. 2016, Gou et al. 2017, Yang et al. 2017, Bao et al. 2018b, Pan et al. 2018), such as the BSR method, the amplitude blanking zone method, the attribute prediction method, the wave impedance prediction method, the saturation prediction method, etc. From the perspective of quantification, these

methods can be roughly divided into three categories: The first category includes the qualitative prediction methods, which are intuitionistic, simple to use and easy to understand, but not conducive to judge the variance of enrichment degree in different locations due to the insufficient quantification. The second category includes the semi quantitative prediction methods, which have certain quantitative characteristics, and some quantitative methods can preliminarily reflect the difference of enrichment degree, which is also relative. The third category includes the quantitative prediction methods, which can absolutely reflect the difference of the degree of hydrate enrichment (Wang et al. 2021). Judging from the current categories of methods for predicting hydrate enrichment, the category of quantitative prediction is the most ideal one. This category includes the methods for predicting wave impedance, elastic impedance, hydrate saturation, etc., among which the method for predicting hydrate saturation is the most intuitive, allowing for a more intuitive reflection of the specific hydrate content in a region. At the same time, this method is critical to obtain the hydrate saturation of an area to evaluate its commercial value. The method of predicting hydrate saturation has developed in tandem with the gradual recognition of hydrate occurrence mode. As early as 1983, Pearson et al. (1983) introduced the Timur equation into studies on hydrate, which includes the idea of calculating hydrate saturation. Subsequently, with the development of experimental technology and the deepening of experimental understanding, it is gradually recognized that hydrate exists in the sea sediment mainly in three modes: suspension mode, particle contact mode and cementation mode (Sun et al. 2003a, Sun et al. 2003b, Hu et al. 2014). Based on different occurrence modes, people have formed different prediction methods, among which, the Timur method of time average equation, Wood modified equation method (the Wood method for short) and the Gassmann method are the classical

ones (Lee et al. 1993, Lee et al. 1996, Song 2003, Wu et al. 2009). Specifically, the Wood method is required for hydrate in suspension mode, and the Timur method is required for hydrate in cementation mode. According to findings of the research carried out by Hu et al. (2014), it can be seen that the hydrate enriched in the Shenhu Sea area exist in suspension mode (Zhang et al. 2007). However, the statistical study on hydrate in the Shenhu Sea area shows that the level of hydrate saturation predicted by the Wood method is quite different from the actual level, and the analysis shows that the systematic error in the application method is caused by the insufficient understanding of sediment composition and porosity in the hydrate layer (Bao et al. 2022). Aiming at the problems existing in the practical application of the Wood method, a prediction method of hydrate saturation called the Wood-SE method is proposed in this paper.

The improved Wood-SE method is mainly divided into four steps to predict hydrate saturation, which are basic data preparation, hydrate saturation calculation before correction, hydrate saturation correction amount calculation and final hydrate saturation calculation. Compared with the Wood method, the research results show that the improved Wood-SE method can reduce the influence of system measurement error and has better prediction accuracy.

Improved method for calculating hydrate saturation

Wood Equation

At present, the Wood method is one of the most commonly used methods for calculating the hydrate saturation in suspension-mode in the sea area (Lee et al. 1993, Song 2003), which is implemented by two equations as shown in formula (1) and formula (2):

$$\frac{1}{\rho_{\rm b}V_{\rm b}^2} = \frac{\phi(1 - S_{\rm h})}{\rho_{\rm w}V_{\rm pw}^2} + \frac{\phi S_{\rm h}}{\rho_{\rm h}V_{\rm ph}^2} + \frac{1 - \phi}{\rho_{\rm m}V_{\rm pm}^2}$$
(1)

$$\rho_{\rm b} = (1 - S)\phi\rho_{\rm w} + \phi S\rho_{\rm h} + (1 - \phi)\rho_{\rm m}$$
⁽²⁾

Where,

 $V_{\rm b}$ = P-wave velocity of hydrate bearing sediments; $V_{\rm pw}$ = P-wave velocity of fluid; $V_{\rm ph}$ = P-wave velocity of pure hydrate; $V_{\rm pm}$ = P-wave velocity of substrate; ϕ = Porosity; $S_{\rm h}$ = Proportion of hydrate in pore space; $\rho_{\rm b}$ = Density of hydrate bearing sediments;

 $\rho_{\rm w} = {\rm Density of fluid};$

 $\rho_{\rm h}$ = Density of pure hydrate; $\rho_{\rm m}$ = Density of substrate.

Calculation of substrate density and P-wave velocity

The substrate is usually not composed of one single substance. Generally speaking, the substrate that forms the seafloor sediments is composed of numerous substances, whose density can be expressed by formula (3):

$$\rho_{\rm m} = \sum_{i=1}^{n} f_i \rho_i \tag{3}$$

Where,

 f_i = Volume percentage of the *i*-th substance;

 ρ_i = Density of the *i*-th substance;

n = Quantity of substances contained in the constituent substrate.

The P-wave velocity of fluid saturated rock can be calculated by formula (4) (Helgerud et al. 1999):

$$V_{\rm pm} = \sqrt{\frac{K_{\rm sat} + \frac{4}{3}G_{\rm sat}}{\rho_{\rm m}}} \tag{4}$$

Where,

 K_{sat} = Bulk modulus of fluid saturated rock;

 G_{sat} = Shear modulus of fluid saturated rock.

The substrate can be regarded as a case of no fluid in the rock, and in such case, the porosity of the rock is 0, while K_{sat} and G_{sat} in formula (4) respectively denotes the bulk modulus and shear modulus of substrate.

The bulk modulus and shear modulus of substrate can be calculated by Hill's average formula, as shown in formula (5) and formula (6):

$$K = \frac{1}{2} \left[\sum_{i=1}^{m} f_i K_i + \left(\sum_{i=1}^{m} \frac{f_i}{K_i} \right)^{-1} \right]$$
(5)

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

Where,

 f_i = Volume percentage of the *i*-th mineral in substrate;

 K_i = Bulk modulus of the *i*-th mineral;

K = Bulk modulus of substrate;

 G_i = Shear modulus of the *i*-th mineral;

G = Shear modulus of substrate.

Calculation of P-wave and S-wave velocity of pure hydrate and sea water

The p-wave velocity and s-wave velocity of a certain substance could be calculated according to young's modulus, density and Poisson's ratio (Lu and

Wang 2011). The specific calculation formulas are shown in formulas (7) and (8). For pure hydrate, the p-wave velocity and s-wave velocity are calculated by formula (7) and formula (8) according to Young's modulus, density and Poisson's ratio of hydrate.

$$V_{\rm p} = \sqrt{\frac{E(1-\sigma)}{\rho(1+\sigma)(1-2\sigma)}}$$
(7)
$$V_{\rm s} = \sqrt{\frac{E}{2\rho(1+\sigma)}}$$
(8)

Where,

E = Young's modulus; ρ = Density; σ = Poisson's ratio; V_p = P-wave velocity; V_s = S-wave velocity.

For the young's modulus and Poisson's ratio of pure hydrate, the volume modulus and shear modulus can be used to calculate them, as shown in Formula (9) and Formula (10).

$$E = \frac{9KG}{3K+G} \tag{9}$$

$$\sigma = \frac{3K - 2G}{2(3K + G)} \tag{10}$$

Where,

K = Bulk modulus of pure hydrate;

G = Shear modulus of pure hydrate.

The P-wave velocity of sea water is generally 1480 m/s. Since the sea water is liquid in which the shear wave cannot propagate, the S-wave velocity in the sea water is 0.

The prediction error of the the Wood method

Suppose that due to the insufficient means of understanding, it is considered that the marine sediment skeleton is composed of 50% of clay minerals and 50% of quartz minerals, while in fact, the seafloor sediment skeleton is composed of three kinds of minerals, namely, 40% of clay minerals, 50% of quartz minerals and 10% of calcite minerals. It is assumed that the porosity of seafloor sediment is 40%, while the actual porosity is 35%, and the hydrate in the pores exists in suspension mode, with the saturation of 30%. In order to facilitate the applicability analysis of the Wood method, the elastic parameters of three minerals used in the comparative analysis are listed in **table I.**

In light of the actual composition of seafloor sediments and judging from **table I**, formula (2) and formula (3), it can be calculated that the density of

the actual hydrate bearing sediments is about 1.97 g/ cm³. Judging from **table I**, formula (3), formula (4), formula (5) and formula (6), it can be calculated that the propagation velocity (V_{pm}) of sesmic P-wave in the skeleton composed of three substances is about 4487.58 m/s. Judging from **table I**, formula (9), formula (10) and formula (7), it can be calculated that the velocity of hydrate bearing sediments is about 1846.89 m/s.

 TABLE I.
 ELASTIC PARAMETERS OF DIFFERENT MIN-ERALS.

Substances	Bulk modulus /GPa	Shear modulus /GPa	Density (g/cm ³)
Calcite	74.4	26.26	2.8
Quartz	40	25.06	2.65
Clay	20.9	6.85	2.58
Sea water	2.5	0	1.032
Pure hydrate	5.6	2.4	0.9

Based on the condition that seafloor sediment skeleton is composed of two substances and judging from table I, formula (5), formula (6), formula (3) and formula (4), it can be calculated that the propagation velocity (V_{pm}) of seismic P-wave in the skeleton composed of two substances is about 4228.73 m/s. According to the density of hydrate bearing sediments, P-wave velocity of pure hydrate and velocity of hydrate bearing sediments calculated previously, the hydrate saturation can be calculated to be about 16.40% by formula (1), which shows that there is a huge error between the calculated hydrate saturation and the real value. However, it is very difficult to eliminate the error caused by the insufficient understanding of substance composition and physical property of sediments, which can be classified as the systematic analytic error of stratigraphic parameters.

Implementation steps of the Wood-SE method

The Wood method has a low prediction accuracy of hydrate saturation due to systematic error. Compared with the Wood method, the proposed Wood-SE method takes the non-hydrate layer overlying hydrate as a reference to obtain the systematic error correction, introduced into the calculation of hydrate saturation to eliminate the systematic error. The implementation steps of this method are divided into the following four steps:

i) Basic data preparation. These data include: (1) Relevant data on elastic parameter, such as mineral composition proportion of substrate, bulk modulus and shear modulus of minerals, density of minerals; bulk modulus and density of sea water; bulk modulus, shear modulus and density of pure hydrate; (2) Relevant data on well (the data must be continuous and include hydrate intervals as well as non-hydrate intervals above hydrates), such as depth, P-wave velocity, porosity and density of sediments.

ii) Calculation of the hydrate saturation before correction. The Wood method is used to calculate the hydrate saturation before correction. Suppose that the hydrate saturation of a certain depth is S_{hi}^{p} , and *i* represents the serial number of depth sampling.

iii) Calculation of hydrate saturation correction. The average value of S_{hi}^{p} calculated by the non-hydrate interval is set to S_{h}^{A} , and then S_{h}^{A} is subtracted from 0 to obtain the difference, which is set to ΔS_{h} .

iv) Final calculation of hydrate saturation. Suppose that the final hydrate saturation is S_{hi}^{t} , then $S_{hi}^{t} = S_{hi}^{p} - \Delta S_{h}$.

Analysis on the application effect of the Shenhu Sea Area

Geological overview of the study area

The study area is located in the Shenhu northern margin of South China Sea (red area in **Fig. 1**), which belongs to Baiyun Sag of Pearl River Mouth Basin in tectonic division (Wu et al. 2009). From April to June 2007, the China Geological Survey (CGS) successively carried out hydrate drilling in places marked with BSR in this area (the position marked with red circle in **Fig. 2**). Hydrate samples were obtained at some drilling positions, such as SH-2, SH-3 and SH-7, whereas hydrate samples were not obtained at some other drilling positions, such as SH-1 and



Fig. 1. Structural location of the research area [revised by Wu et al. (2009)].

SH-4 (Zhang et al. 2014). Three wells with hydrate samples are located in the ridge of rugged seafloor of continental slope with a depth of 1200 m, and the obtained gas hydrate samples are mainly distributed in the sediment pores about 200 m below the seafloor (Chen et al. 2018). The composition of sediments near hydrate samples can be divided into three types according to the size of particles (Chen et al. 2011): (1) Sand, which refers to particles with diameter of more than 0.063 mm; (2) silty sand, which refers to particles with diameter of 0.004-0.063 mm; (3) clay, which refers to particles with diameter of less than 0.004 mm. The main type of particles in sediments is silty sand, followed by clay and sand, among which the average content of silty sand is about 75%, that of clay is about 20%, and that of sand is about 5%.



Fig. 2. Topographic features of hydrate sampling area in the Shenhu Sea area, Northern South China Sea [revised by Wu et al. (2009)].

In terms of the porosity performance law of seafloor sediments in this area, in 2011, Guo et al. (2011) conducted laboratory analysis on the porosity of drilling samples in the Shenhu Sea area, and found that there is an approximate relationship between the porosity and the depth of seafloor sediments, as shown in formula (11):

$$\phi = -7.44 \times \ln D + 85.38 \tag{11}$$

Where, $\phi = Porosity, \%;$ D = Depth of sediments under the seafloor, with the unit of m.

Acoustic logging curve of the well S and the hydrate saturation calculated by chloride ion

Figure 3 and **figure 4** show the curves of P-wave velocity and density of one of the three wells with drilled hydrate (assuming that this well is the well S). It can be seen from the acoustic logging curve of the well S, that when the depth is between 200-220 m, the P-wave velocity increases, reaching 2300 m/s at the peak level, while the density decreases, thus this position has been confirmed to contain hydrate (Liang et al. 2010).

During the formation and decomposition of hydrate in the sea area, the concentration of chloride ions will change, repelling the salt ions dissolved in pore water during the formation, leading to the diffusion of salt ions outwards. Whereas when the hydrate decomposes, the pore water will be diluted, leading to the lower salinity of the samples with hydrate obtained by drilling than those without hydrate. Hydrate with different saturation often has different



Fig. 3. The variation of P-wave velocity with depth in the well S.



Fig. 4. The variation of the well density with depth in the well S.

concentration of chloride ions, so the hydrate saturation can be calculated according to the concentration of abnormal chloride ions in the pore. Some hydrate saturation of the well S obtained on the basis of abnormal concentration of chloride ions in hydrate is shown in **figure 5** (Zhang et al. 2007). Judging from the comparison of **figure 3** and **figure 5**, at the depth where the P-wave velocity is relatively large, the hydrate saturation is also relatively large, which can reach more than 30%, with a maximum of about 50%.

Calculation of elastic parameters of the skeleton

Referring to the above analysis on the composition of particle sizes of hydrate-bearing sediments, the proportions of particle size composition used to calculate sediment skeleton are set as follows: 75% for silty sand, 20% for clay and 5% for sand. The bulk modulus, shear modulus and density data of siltstone, sandstone and pure methane hydrate are listed in **table II** (Liang et al. 2010, Bao et al. 2018a). Since the particle sizes of silty sand and sand in the study area include different diameters, it is complicated to accurately describe their elastic parameters. To facilitate



Fig. 5. Estimates of hydrate saturation in the well S based on concentration of chloride ions.

 TABLE II. ELASTIC PARAMETERS OF VARYING SEDI-MENT COMPONENTS.

Substances	Bulk modulus /GPa	Shear modulus /GPa	Density /(g/cm ³)
Silty sand	40.08	28.13	2.65
Sand	40	25.06	2.65
Pure methane hydrate	5.6	2.4	0.9

the study, the elastic parameters of siltstone (formed by silty sand of varying particle sizes) and sandstone (formed by sand of different particle sizes) in **table II** will be used to approximately replace the elastic parameters of silty sand and sand in the study area.

The skeleton of sediments consists of three particle size components. According to formula (5) and formula (6), the bulk modulus and shear modulus of the skeleton can be shown in formula (12) and formula (13).

$$K = \frac{1}{2} \left[\sum_{i=1}^{3} f_i K_i + \left(\sum_{i=1}^{3} \frac{f_i}{K_i} \right)^{-1} \right]$$
(12)

$$G = \frac{1}{2} \left[\sum_{i=1}^{3} f_i G_i + \left(\sum_{i=1}^{3} \frac{f_i}{G_i} \right)^{-1} \right]$$
(13)

Where,

 f_1 = Volume percentage of siltstone in the skeleton; f_2 = Volume percentage of sandstone in the skeleton;

- f_3 = Volume percentage of clay in the skeleton;
- K_1 = Bulk modulus of siltstone;
- K_2 = Bulk modulus of sandstone;
- K_2 = Bulk modulus of clay;
- G_1 = Shear modulus of siltstone;
- G_2 = Shear modulus of sandstone;
- G_3 = Shear modulus of clay.

According to the data in formula (12), formula (13) and **Table I**, it can be calculated that the bulk modulus and shear modulus of the skeleton are approximately 35 GPa and 20.5 GPa respectively.

The density of the skeleton can be calculated by formula (14).

$$\rho_{\rm m} = f_1 \rho_1 + f_2 \rho_2 + f_3 \rho_3 \tag{14}$$

Where,

 ρ_1 = Density of siltstone;

 ρ_2 = Density of sandstone;

 ρ_3 = Density of clay.

According to the data in formula (14) and **table I**, it can be calculated that the density of the skeleton is 2.64 g/cm^3 .

Comparison and analysis of prediction results of both methods

The hydrate saturation measured by the chloride ion method is considered to be a reliable measurement result at present. This paper takes the results measured by this method as the standard for the experimental evaluation.

The comparison of hydrate saturation in the well S predicted by the the Wood method and the Wood-ES method with hydrate saturation calculated by the chloride ion method is shown in **figure 6**, where the dotted line represents the prediction result of the Wood method, the blue solid line represents the prediction result of the Wood-ES method, and the blue circle represents the calculation result of the chloride ion method. It can be seen from figure 6 that the hydrate saturation predicted by the Wood method at different depths is quite different from that calculated by the chloride ion method. Generally, the absolute value error is more than 20%, and even more than 40% in some positions (Kang et al. 2018, Wang et al. 2019, Zhang et al. 2019). However, for the Wood-ES method, when the sediment depth is less than 200 m, the hydrate saturation predicted by this method is basically the same as that calculated by the chloride ion method (Zhang et al. 2006). When the sediment depth is between 200-220 m, the hydrate saturation predicted by this method is in the high-value region, and most of the predicted values are smaller than



Fig. 6. Comparison of hydrate saturation predicted by the Wood method and the Wood-ES method with hydrate saturation calculated by the chloride ion method.

those calculated by the chloride ion method, though the difference is not significant (Chen et al. 2018, Han et al. 2020). When the depth is greater than 220 m, the hydrate predicted by this method is larger than that calculated by the chloride ion method.

The comparison of absolute error (referred to as prediction error) predicted by the two methods at different depths is shown in **Figure 7**, which shows that the prediction error of the Wood method falls within the range of -10%, 60%, while that of the Wood-ES method mostly falls within the smaller range of -10%, 10%, which shows that the Wood-ES method is obviously superior to the Wood method. Because the Wood-SE method has good prediction accuracy, it has great application prospect in saturation prediction of hydrate layer with suspension mode similar to Shenhu Sea Area.

CONCLUSIONS

Based on the systematic study of the methods for predicting hydrate saturation in suspension mode in the sea area, the following conclusions are reached:



Fig. 7. Comparison of absolute error between the Wood method and the Wood-ES method in different depths.

- (i) In this paper, a method of predicting hydrate saturation to eliminate systemic error is proposed, namely the Wood-ES method. Compared with the Wood method, the Wood-ES method can better address the issue of large prediction error caused by the difference between actual measurement and theoretical hypothesis, which is beneficial to improve the effect of prediction method in practice.
- (ii) Taking the actual data collected from the Shenhu Sea Area in China as an example, we have carried out a comparative study on the prediction of hydrate saturation by the two methods. It is pointed out that the Wood-ES method is a rather good method for predicting hydrate saturation.
- (iii) The Wood-ES method to eliminate systemic error will have higher value of popularization in practice thanks to its smaller prediction error.

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