

## Calculation and applied analysis of natural gas hydrate saturation based on experimental data

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The saturation is a very important parameter to evaluate natural gas hydrate. In order to accurately calculate the natural gas hydrate saturation during the synthesis process and analyses the relationship between resistivity and saturation in hydrate deposits, the resistivity and pressure values during the synthesis of natural gas hydrate were measured. Based on the instrument calibration and gas equation of state, the calculation model of natural gas hydrate saturation is established. The relationship between the saturation value of natural gas hydrate and the resistivity is discussed under different particle size, compaction and shale content. The results show that there is a certain exponential relationship between the saturation of gas hydrate and resistivity and a nonlinear relationship between gas hydrate saturation and resistivity index. There are some differences from the relationship in Archie's formula. Therefore, the Archie formula is not suitable for the calculation of hydrate saturation in sediment by logging data. Through the application analysis, it is concluded that the experimental model of gas hydrate saturation has the advantages of high accuracy and applicability.

Keywords: Gas hydrate Saturation; Rock physical experiment; P-V-T equilibrium equation; Resistivity Index

### INTRODUCTION

Gas hydrate is the cage-type crystalline compounds formed under high pressure by water and natural gas in low temperature; the interaction between water molecules makes the stable solid lattice and gas molecules occupy the body cavity of lattice [1, 2]. Gas hydrate of nature is mainly distributed in the Marine sediments of the continental slope and the frozen continent; it is estimated that the organic carbon content in gas hydrate is twice as the other traditional resources all over the world such as a recoverable natural gas and oil [3, 4]. Gas hydrate is praised as new types of potential energy which is clean, efficient and rich reserves because of widely distributed, rich reserves, high density, etc [5]. And it also has been given high attention and researched in many countries [6, 7].

Archie formula, Dual water model and some other formulas like these have been used to calculate the gas hydrate saturation [8-12]. Tinivella estimated gas hydrate saturation according to the longitudinal wave with the theory of application of double phase medium [13]; Lee pointed out that weighted of three-phase time average equation and the three-phase Wood equation can be used to estimate deep sea sediment velocity of hydrate [14,15]; Wang Xiujuan estimated gas hydrate saturation with the application of P-wave velocity thermal elastic theory and considered the influence of the thermodynamic

properties to saturation estimation results, which is effective [16]; Mo Xiuwen, on the basic of previous studies, put forward the method that is using of logging interpretation of chloride ion mass concentration to calculate the hydrate saturation [17]; Chen Yufeng studied the relationship between the hydrate saturation and resistivity with the using of natural gas hydrate resistivity measurement system. And the results showed that increase resistivity index and water saturation are not like linear relationship which is described by Archie formula, it considered that the resistivity calculated saturation requires correction [18, 19].

This article uses SHW-III acoustic power test device of gas hydrate which is with high visualization degree, complete measurement data and real-time data monitoring, and uses the method of continuous supply of natural gas in stages to the greatest extent on the synthesis of gas hydrate. Using P-V-T equilibrium equation calibrate the related parameters of the hydrate instrument in the first place, and then deduced the theory of calculation of natural gas hydrate saturation equation, to achieve the purpose of accurate calculation of natural gas hydrate saturation finally. Verification and analysis the calculation method of gas hydrate saturation through the experimental data.

### SATURATION CALCULATION

Natural gas hydrate is mainly formed by continuous supplication of natural gas in stages, so

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the volume of measuring room and pipeline in experimental device will have a greater impact on the calculation result. Therefore, it is necessary to calibrate the volume of the measuring room  $V_1$  and pipeline  $V_s$ , the schematic diagram of experimental device shown in Fig. 1.

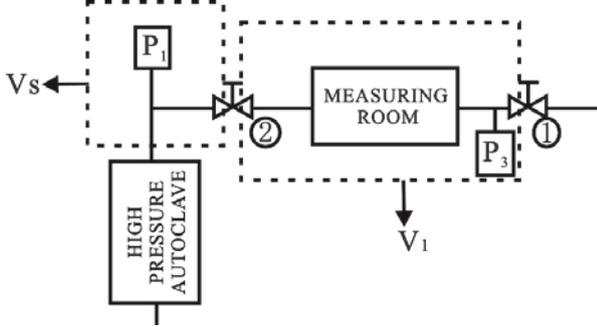


Fig. 1. Experimental device schematic diagram

Calibrate the volume of the measuring room  $V_1$  and pipeline  $V_s$ : Firstly, put the solid stainless steel module into autoclave, fill with a certain pressure of gas in the system, and then close the valve ② and open the valve ①. Next, empty the gas of measuring room and record the initial stable pressure  $P_{1A}$ ; And then close the valve ①, open the valve ② and record the pressure for a steady state  $P_{1B}$ . At this point establish the P-V-T equation in balance state:

$$\frac{P_{1A} V_s}{Z_{1A}} = \frac{P_{1B} (V_s + V_1)}{Z_{1B}}$$

Where  $P_{1A}$  is the initial stable pressure,  $P_{1B}$  is the steady state pressure,  $Z_{1A}$  is the compression factor of gas in the pressure  $P_{1A}$  for conditions,  $Z_{1B}$  is the compression factor of gas in the pressure  $P_{1B}$  for conditions.

Put a module with known volume  $V_2$  into measuring room and repeat the above steps. Record the steady state pressure  $P_{2A}$  and  $P_{2B}$  when the valve ② closing and opening. Meanwhile establish the P-V-T equation in balance state:

$$\frac{P_{2A} V_s}{Z_{2A}} = \frac{P_{2B} (V_s + V_1 - V_2)}{Z_{2B}}$$

Where  $P_{2A}$  is the initial stable pressure,  $P_{2B}$  is the steady state pressure,  $Z_{2A}$  is the compression factor of gas in the pressure  $P_{2A}$  for conditions,  $Z_{2B}$  is the compression factor of gas in the pressure  $P_{2B}$  for conditions.

By the above two P-V-T equilibrium equation of state the measuring room volume  $V_1$  and volume of pipeline  $V_s$  can be obtained. In order to improve the accuracy, changing different volume of modules load to measuring room and repeat the processes can get more P-V-T equilibrium equations under the condition of constant temperature, with the

simultaneous Formula 1 multiple of the measuring room volume  $V_1$  and the volume of pipeline  $V_s$  can be calculated, and final averaged.

The formation of gas hydrate is mainly adopts the method of continuous supplication of natural gas in stages which has the following steps: firstly, instead of certain water put amount of gas into porous medium filled with water, to make a suitable space for the formation of hydrate; Then continuing in stages to pass into the gas in porous media, synthesis of hydrate and calculation of water saturation and gas hydrate saturation at equilibrium under appropriate conditions, repeating the above process until the water in the porous media is completely consumed. Assuming  $V_{e_i}$ ,  $V_{w_i}$ ,  $V_{c_i}$  respectively the volumes of gas, residual water and hydrate in porous media for  $i$  time at the start of the reaction synthesis. And the synthetic process of natural gas hydrate can be expressed in Fig. 2.

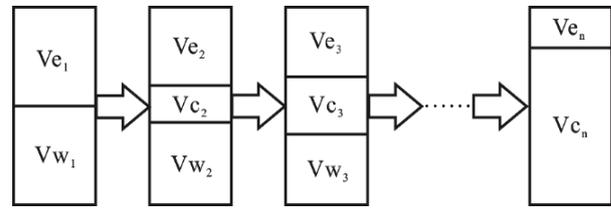


Fig. 2. The synthetic process diagram of natural gas hydrate

As take the first natural gas hydrate synthesis process as an example: at the start of the reaction, the inner pressure of the autoclave is  $P_{1a}$  and the corresponding compression factor is  $Z_{1a}$ , P-V-T equilibrium equation can be established when system for the amount of substance  $n_{1a}$  gas filled the space of autoclave  $V_{e1}$  and the volume of pipeline  $V_s$ .

$$P_{1a} (V_{e1} + V_s) = Z_{1a} n_{1a} RT$$

At the end of the reaction, the pressure in the autoclave is  $P_{1b}$  and the corresponding compression factor is  $Z_{1b}$ , P-V-T equilibrium equation can be set up when system for the amount of substance  $n_{1b}$  as filled the space of autoclave  $V_{e2}$  and the volume of pipeline  $V_s$ .

$$P_{1b} (V_{e2} + V_s) = Z_{1b} n_{1b} RT$$

The consumption of natural gas amount of substance in the synthesis reaction process of natural gas hydrates of the first time is:

$$\Delta n_1 = n_{1a} - n_{1b} = \frac{P_{1a} (V_{e1} + V_s)}{Z_{1a} RT} - \frac{P_{1b} (V_{e2} + V_s)}{Z_{1b} RT}$$

The volume of natural gas  $Ve_1$  in autoclave can be calculated by the water discharged at the first time. And  $Ve_2$  can be calculated by the following processes:

Put the amount of substance  $n_3$  in the measuring room so that the stable pressure is  $P_3$  and the corresponding compression factor is  $Z_3$ . At the same time, P-V-T equilibrium equation 6 can be established.

$$P_3V_1 = Z_3n_3RT$$

P-V-T equilibrium equation can be established after opening valve ② so that the part of the natural gas spread into autoclave and expanded rapidly to reach equilibrium. Meanwhile, the pressure becomes  $P_{2a}$  and corresponding to the compression factor becomes  $Z_{2a}$ .

$$P_{2a}(Ve_2 + V_s + V_1) = Z_{2a}(n_{1b} + n_3)RT$$

By the above two P-V-T equilibrium equations, we can get the expression of  $Ve_2$ :

$$Ve_2 = \left[ \frac{P_{2a}(V_s + V_1)}{Z_{2a}} - \frac{P_{1b}V_s}{Z_{1b}} - \frac{P_3V_1}{Z_3} \right] \left/ \left[ \frac{P_{1b}}{Z_{1b}} - \frac{P_{2a}}{Z_{2a}} \right] \right.$$

The use of the above formula can be calculated natural gas consumption  $\Delta n_1$ , and according to natural gas hydrate formation reaction equation calculates the corresponding water consumption of natural gas consumed  $\Delta n_1$  moles, as expressed by volume  $\Delta Vw_1$ . At this point the remaining water volume in the autoclave is:

$$Vw_2 = Vw_1 - \Delta Vw_1$$

Where can be obtained based on the calculation of quality of the first water discharged. Thus, when the end of the first synthesis gas hydrates, water saturation of the porous medium is:

$$S_w = \frac{Vw_2}{V_\phi}$$

Gas hydrate saturation is:

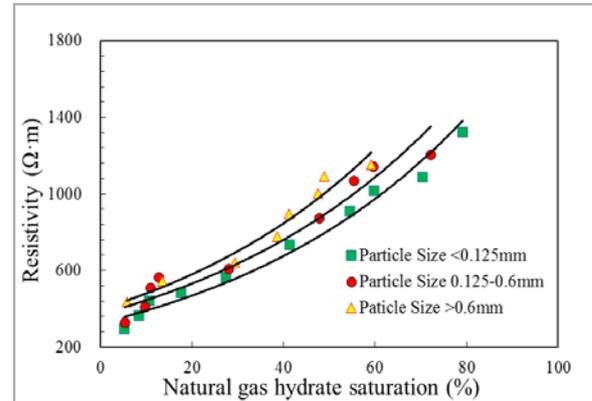
$$S_c = \frac{Vc_2}{V_\phi} = \frac{V_\phi - Vw_2 - Ve_2}{V_\phi}$$

According to the above steps, the water saturation and the hydrate saturation value in the porous medium at the end of the hydrate formation at each stage can be obtained.

## LITHO-ELECTRIC RELATION ANALYSIS

According to the records of experimental parameters and combining with the theory on the calculation formula, water saturation, gas saturation, hydrate saturation and resistivity and resistivity index and so on in natural gas hydrate formation in porous medium in the autoclave are all obtained with the condition of reaction temperature is 1°C.

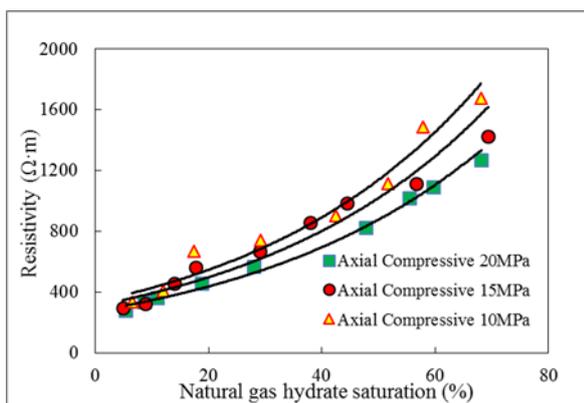
Under the experimental condition that the reaction temperature is for 1°C, the relationship between resistivity and the hydrate saturation in different particle size、different axial compressive and different shale content of porous medium to form natural gas hydrate in autoclave as shown in Figure 3、Figure 4 and Figure 5. Under the condition of three different particle size、three different axial compressive and three different shale content of rock sample text, the calculations obtained hydrate saturation ( $S_c$ ) and resistivity ( $R$ ) in linear coordinates show a certain exponential relationship from Fig. 3、Fig. 4 and Fig. 5; With the larger grain size, lowered cementation degree and decreasing the shale content, the resistivity increase when containing the same natural gas hydrate saturation.



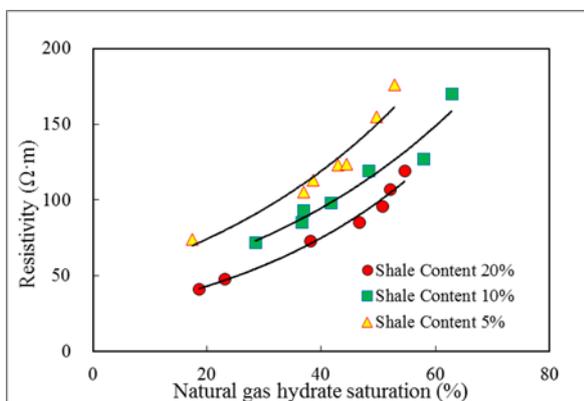
**Fig. 3.** The relationship between hydrate saturation and resistivity in different particle size experimental conditions

The relationship between resistivity index ( $I$ ) and water saturation ( $S_w$ ) in the natural gas hydrate formation process in porous media with the condition of reaction temperature is 1°C in the autoclave as shown in Fig. 6、Fig. 7 and Fig. 8. The figures show that in the linear coordinates the relationship of resistivity index ( $I$ ) and water saturation ( $S_w$ ) from three different particle size、three different axial compressive and three different shale content of sample test is not a line (but there exists similarities of change trend). It means

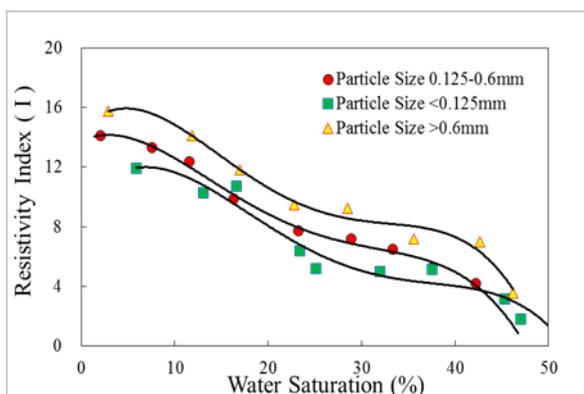
that under the experimental conditions, hydrate sediments have the phenomenon of Non-Archie.



**Fig. 4.** The relationship between hydrate saturation and resistivity in different axial compressive experimental conditions



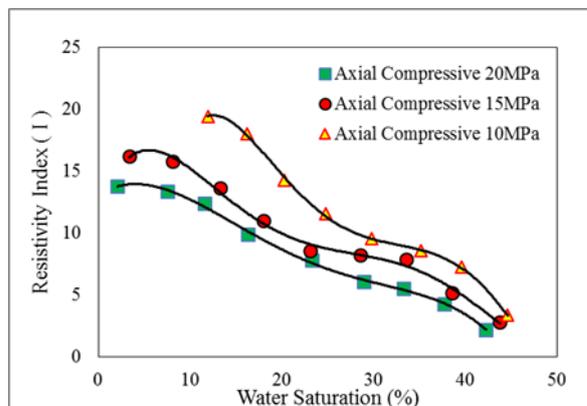
**Fig. 5.** The relationship between hydrate saturation and resistivity in different shale content experimental conditions



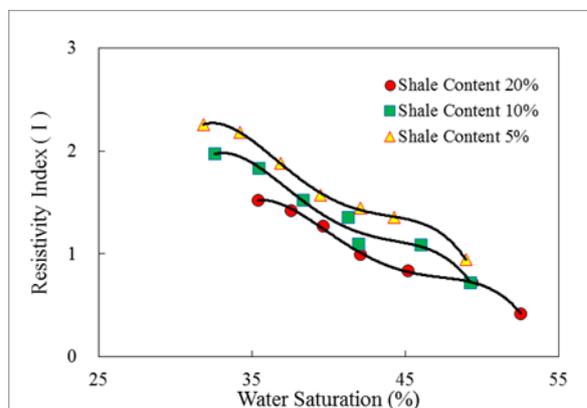
**Fig. 6.** The relationship comparison chart of resistivity index and water saturation in different particle size experimental conditions.

That because Archie formula is only suitable for pure sandstone. We change the state of pure sandstone and size of original pore after the synthesis of natural gas hydrate in porous media so that there is Non-Archie phenomenon existing between resistivity index ( $I$ ) and water saturation

( $S_w$ ). It is the same with the conclusion from Yufeng Chen in the research of the relationship between sediment gas hydrate saturation and resistivity [11].



**Fig. 7.** The relationship comparison chart of resistivity index and water saturation in different axial compressive experimental conditions



**Fig. 8.** The relationship comparison chart of resistivity index and water saturation in different shale content experimental conditions

## CONCLUSION

1. Through this experiment, we formed a new natural gas hydrate synthesis method, a set of instrument-related parameters calibration method and a set of synthetic gas hydrate process of natural gas hydrate saturation computational model.
2. The data analysis results show that: the saturation of natural gas hydrate ( $S_c$ ) and the resistivity( $R$ ) shows a certain exponential relationship, the water saturation ( $S_w$ ) and the resistivity index ( $I$ ) did not show the linear relationship (have the phenomenon of Non-Archie); With the larger grain size, lowered cementation degree and decreasing the shale content, the resistivity increase when containing the same natural gas hydrate saturation.
3. The results of comparative analysis show that: The result of this experiment accord to the objective phenomena and laws of rock physics experiments, so

the method adopted to synthesize the natural gas hydrate and the computational model is correct in the experiment, which put forward a new idea computing the saturation of natural gas hydrate.

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