# COMPRESSIONAL WAVE RESPONSE OF METHANE HYDRATE FORMATION IN BEARING SEDIMENTS

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# ABSTRACT

A system for methane hydrate-bearing sediment detection with pressure is designed, series of experiments for compressional wave response of hydrate-bearing sands are performed systematically in laboratory. Considering the difficulties in performing valid laboratory tests and in recovering intact hydrate bearing sediment samples. The system is designed to achieve in situ hydrate formation in bearing sediments and synchronous acoustic detection and signal analysis. The hydrate formation unit comprises a pressure chamber which is designed for pressure core analysis, a high-precision pump for gas injection. The ultrasonic test unit consists of a pulse transceiver, an A/D converter, an ultrasonic transducer, a signal amplifier and an oscilloscope. The influence factors, including hydrate saturation and formation process to P-wave velocity (Vp) and amplitude are investigated. The results show that Vp and amplitude both increase with the hydrate saturation. Vp exhibit relatively minor changes when hydrate saturation varies from 0% to 40% compared to the P-wave amplitude. However, there is a close relationship between the velocity of P-wave and the saturation of hydrate which could be the effective criterion for the hydrate formation in bearing sediments.

**Keywords:** ultrasonic test; gas hydrate sediment; P-wave velocity; hydrate saturation

# 1. INTRODUCTION

As a kind of clean energy with high energy density and large energy reserves, natural gas hydrate has become a potential alternative energy resource. At present, the exploration and development of natural gas hydrate in the world has promoted lots of researches and technologies. With the development of natural gas hydrate sampling techniques, detection and analysis of gas hydrate pressure cores become the bridge connecting hydrate exploration and exploitation. Furthermore, sampling techniques of pressure cores reflect the reservoir characteristics and hydrate accumulation which is important for the study of hydrate exploitation.

Remotely estimating hydrate saturations in the natural environment requires accurate seismic velocity information for hydrate-bearing sediments. W.F. Waite [1] presents a set of experiments to study unconsolidated, partially water-saturated Ottawa sand samples containing an interconnected methane gas phase. T.S. Yun [2] clarifies the microscale processes accompanying hydrate formation in porous media and provide guidelines for the extrapolation of laboratory results to field settings. Laboratory measurements of compressional wave and shear wave velocities in hydrate-bearing sands imply that hydrate formation primarily affects the bulk stiffness of sediments, a result consistent with the pore filling hypothesis.

However, the formation of gas hydrate is usually at high pressure and low temperature environment. As is well known that acoustic propagation in different medium is not the same, the ultrasonic detection outside the pressure maintaining transfer device will face the problem of wave diffraction. Therefore, special system must be designed to meet the detection requirements. This study presents a novel ultrasonic test system for pressure conserved methane hydrate-bearing sediment, this test system consists of two main units, which can be used both for formation of hydrate at low temperature and for external P-wave measurements at in situ

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pressures. The P-wave responses can be used to predict the hydrate saturation and morphologies in the bearing sediments. The P-wave measurements provide a preliminary understanding of natural cores, which is significant for the subsequent exploration and later detailed core analysis in a laboratory.

## 2. EXPERIMENTAL DESIGN

## 2.1 Apparatus and materials



Figure 1. Diagram of the experimental system

In this experimental system, a special design of the chamber is carried out according to the requirements of the fast detection of the methane hydrate-bearing sediment, and the device structure is shown in Figure 1. Similar to the apparatus used in the previous study [3], a J-typethermistor is used in this system to monitor the temperature change. The design of the details derives from our unique technique; a fiberglass pipe is used to enhance the mechanical strength. The main function of this unit is to generate the high-pressure and lowtemperature conditions for the formation of hydrates. The ultrasonic test unit includes a pulse transceiver, an ultrasonic transducer, an A/D converter, a signal amplifier and an oscilloscope. The ultrasonic pulsereceiver offers square wave excitation which is especially powerful in testing highly attenuating materials. Ultrasonic waves of different frequencies have different propagation characteristics in sediments. The ultrasonic signal has a strong resolution with high frequency and short wavelength, however the signal strength is easy to attenuate rapidly in the sediment sample. When the ultrasonic frequency is relatively low and the wavelength is longer. The ultrasonic signal intensity is relatively strong which can penetrate the thicker sediment sample, however the relative signal resolution is decreased. Therefore, it is of great significance to choose the appropriate ultrasonic frequency for measurement. In order to weight the advantages and disadvantages, the

frequency of the ultrasonic transducers used in the experiment is 100 kHz. The received signal is digitized, amplified and then displayed on the digital phosphor oscilloscope (DPO 4034B, Tektronix, Inc., USA). The dynamic variation of the waveform is recorded by the computer for further analysis.

The ultrasonic transducers are fixed to the polymer protecting jackets which are designed to avoid wave diffraction and to withstand high pressure. Additionally, the protecting jackets also function as a matching layer that increases the transmission power of ultrasonic waves between the transducers in terms of ultrasonic wave attenuation during propagation. Each polymer protecting jacket is embedded into a round hole with a diameter of 44mm which penetrates through the side wall of the chamber. The protecting jackets and the wall must be completely sealed to ensure the pressure stability in the chamber. The chamber is made of stainless steel with a wall thickness of 20 mm, length of 300 mm and an inside diameter of 80 mm. The dimensions are specially designed to withstand the high operating pressure (20 MPa).

Quartz sands are used to investigate the impacts acoustic response on the saturation of gas hydratebearing sediments. Deionized water and methane gas with a minimum purity of 99.0% are injected into the reaction cell at a specific temperature and initial pressure for formation of the hydrate samples.

## 2.2 Experimental procedures

In the study of the acoustic characteristics of methane hydrate-bearing sediment, the detailed experimental process is as follows.

(1) Before the experiment starts, the deionized water is used to clean the chamber at least 3 times.

(2) The clean and dry quartz sands are first packed into the vessel with a certain amount of deionized water saturating pore spaces.

(3) Three pairs of ultrasonic transducers are assembled on the protecting jacket together with a pressing cap and several lock screws. To reduce acoustic energy attenuation, an ultrasonic couplant is smear evenly on the surface of each ultrasonic transducer. All components of the ultrasonic test unit are connected by BNC (Bayonet Nut Connector) cables and debugged in advance.

(4) After injecting methane gas at a preset pressure, the chamber is placed in an air bath at 274 K for hydrate formation.

(5) With sensors monitoring the pressure and temperature changes, the formation process is

considered completed when the pressure sharply dropped and then stabilized again for at least 2 h. The acoustic responses of the methane hydrate-bearing sediment are displayed on the oscilloscope and the data is collected by the computer. The data are analyzed to estimate hydrate saturation.

# 2.3 Theoretical methods

In this paper, acoustic transmission method is used to measure the P-wave velocity of hydrate-bearing sediment, three pairs of ultrasonic transducers are employed for transmitting and receiving ultrasonic signals. The propagation time of the ultrasonic wave in the sample is the first arrival time of the receiving signal. The propagation distance L of the ultrasonic wave is the summation of the bearing sediment and the protecting layer. P-wave velocity of hydrate-bearing sediment is determined by Equation (1):

$$V_p = \frac{L}{t - t_0} = \frac{L}{t_p}$$

The first wave arrival time can be obtained according to the waveform obtained by the oscilloscope. During the experiment, the ultrasonic transducer is not directly in contact with the bearing sediment, so it is necessary to measure the system delay before calculating P-wave velocity. The delay time is related to the internal structure of the transducers which can be read from the oscilloscope directly by connecting the transmitter and receiver.

## 3. RESULTS AND DISCUSSION

#### 3.1 Discussion

Figure 2 shows the change of temperature and pressure during methane hydrate formation. Combined with the temperature and pressure curve, at about 190 min, the temperature rises and the pressure drops abruptly, which proves that the hydrate starts to generate. Correspondingly, the P-wave velocity of the sediment is significantly increased, indicating that the presence of hydrate can change the elastic properties of the bearing sediment. Then, during 190 ~ 300 min range, pressure curve decreases slowly and hydrate formation continues, P-wave velocity can effectively reflect a range of hydrate saturation values.



Figure 2. The change of temperature and pressure during methane hydrate formation

## 3.2 Dynamic Waveforms

Figure 3 illustrates the waveforms during the process of methane hydrate formation which only shows the acoustic signals of 285 min, 350 min and 625 min for simplification. It can be seen from the diagram, as the change of time, acoustic amplitude changes significantly after the methane hydrate formation, especially at the late stage of formation, the wave amplitude is much greater than the early stage of hydrate formation. At 285 min, the amplitude of the acoustic wave is enhanced, and the effect of hydrate on the sediment began to appear. At about 350 min, the amplitude of the acoustic wave tends to be stable, and at the end of 625 min, the amplitude of the acoustic wave remains the maximum.



Figure 3. Acoustic signals of 285 min, 350 min and 625 min during methane hydrate formation

3.3 Effects of hydrate saturation on P-wave velocity



Figure 4 shows the waveform diagram of the sediment samples with different hydrate saturation under the condition of excessive water. The hydrates formed are suspended in the pore water and keep a uniform distribution at a low level hydrate saturation. At this moment, the existence of hydrate could not change the mechanical properties of the sediment, which is shown as the stability of the P-wave velocity. At the middle stage of hydrate formation, hydrates are aggregated and attached to the surface of sediment particles leading to the increase of the attenuation. When hydrate particles gradually unite or bound to particles, they begin to affect the elastic properties of the whole sediment, which is manifested as a significant increase in P-wave velocity. Similarly, at the late stage of hydrate formation, hydrate layers covering the surface of particles are gradually thickened. The penetration efficiency of acoustic waves is further improved, but the P-wave velocity is not sensitive to the thickening of hydrate layers.

Figure 5 illustrates the conceptual diagram showing different pore-fluid displacing hydrate morphologies. Pore-fluid displacing hydrate can be subdivided into cementing or non-cementing morphologies based on whether hydrate grows adhering to sediment grains or floating in the pore fluid inside the pore space. The distinction between different pore-fluid displacing hydrate morphologies is effectively deduced from the elastic wave velocity. The cementing morphology has a much greater effect on the elastic properties of hydratebearing sediments than the non-cementing morphology. Therefore, it could be predicted that pore-filling hydrate mainly concentrates in the water-rich seabed stratum at low saturation, and gradually changes into load-bearing or even cementation type with increasing saturation as shown in Figures 5a and b.



Figure 5. Conceptual diagram showing different pore-fluid displacing hydrate morphologies.

## 4. CONCLUSION

This paper carries out a series P-wave analysis on the formation of excess water samples. The P-wave velocity measurements are shown to provide a preliminary understanding of the acoustic properties of the pressure core samples. The experimental results show that the P-wave velocity is significantly increased in the initial stage of hydrate formation. When the saturation further increases, the hydrate shell gradually thickens, but the P-wave velocity does not change significantly. In other words, the evolution of P-wave velocity is not sensitive to the formation method when the hydrate is in the high saturation. It is also proved that the transformation of acoustic properties is mainly caused by the vibration conduction mode between hydrate and sediment particles.

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