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Acoustic properties of hydrate-bearing sand samples: laboratory measurements (*setup, methods, and results*)

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Abstract

A new system has been designed for laboratory physical modeling of hydrate-bearing sand samples and measuring their acoustic properties at different temperatures and pressures. The system includes a pressure vessel, units of temperature control, external pressure, and gas/liquid delivery, and a unit for measuring velocities of acoustic waves. Measurements are carried out in 10–50 mm high cylindrical specimens 30 mm in diameter. The system provides methane hydrate formation in sand samples and their acoustic measurements for as long as several days due to automatic control. Hydrate-bearing samples are prepared by pressurized methane injection into pores of wet sand and are exposed to several cooling/heating cycles to increase hydrate formation rates. Hydrate-bearing samples have been prepared and travel times of acoustic *P* and *S* waves have been measured in dozens of successful experiments. Acoustic data confirm the formation of hydrates, with the related increase in wave velocities to values about those in frozen sediments. The prepared gas hydrates are inferred to be of “cementing” type, i.e., they form as cement at the boundaries of mineral grains. The obtained velocities of acoustic waves show a positive linear correlation with hydrate contents in sand samples.

Keywords: hydrate-bearing samples; laboratory acoustic measurements; acoustic properties of hydrate-bearing samples

Introduction

Hydrates of hydrocarbon gases, mainly methane, are of broad natural occurrence. They can form and keep stable under certain pressures (*P*) and temperatures (*T*), in the presence of sufficient amounts of water and hydrate forming gas, free or dissolved to a certain concentration (Istomin and Yakushev, 1992; Sloan, 2003). These conditions exist in deep-water marine or lake sediments and in permafrost. Most of gas hydrate deposits occur in unconsolidated bottom sediments and are fed from active submarine sources of fluids and gases under water depths below 300–500 m (Makogon et al., 2007; Mazurenko and Soloviev, 2003): in the Black, Okhotsk, and Caspian Seas, and in Lake Baikal (Ginsburg and Soloviev, 1994; Khlystov, 2006; Obzhirov et al., 2012). Gas hydrates are expected to occur in Arctic seas and their remnants may be found also in permafrost of Siberia (Yakushev, 2009).

Gas hydrates is a potentially rich unconventional source of gas, in amounts of ~2×10^{14} m^3 at 200–300 m below the sea floor and of the order of 10^{13} m^3 near it, according to most realistic estimates by Soloviev (2003). Test production of gas hydrates has been performed successfully from the Mallik deposits in permafrost of the Mackenzie delta, northern Canada, in 2002–2008 (Dallimore et al., 2008; Numasawa et al., 2008) and from submarine deposits in the area of the Nankai trough east of the Japan islands in 2013 (Yamamoto, 2014). Accumulations of gas hydrates influence the stability of structures and offshore wells set on the sea floor. Methane from hydrate deposits may be involved into global climate processes, and special technologies are being developed for sequestering industrial carbon dioxide in hydrated form in permafrost or seafloor settings.

Much effort has been undertaken recently for exploration of gas hydrates, mainly in offshore environments (Akselrod,

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2009). Geophysical surveys by seismic and resistivity methods provide efficient exploration tools due to correlation between the physical properties of sediments and gas hydrate contents. The properties of pure methane hydrate have been quite well known (Sloan and Koh, 2008), while the thermal, electrical, and acoustic properties of hydrate-bearing sediments are more difficult to study and are currently a subject of laboratory research with special systems for preparing such samples (Gabitto and Tsouris, 2010; Riedel et al., 2010; Waite et al., 2009). There exist several instruments of this kind, like the GHASTLI system (Gas Hydrate And Sediment Test Laboratory Instrument) designed in the USA (Winters et al., 2000) which measures acoustic, electrical, and mechanic properties of samples during the formation or dissociation of gas hydrates inside them. Another example is the FLECAS system (Field Laboratory Experimental Core Analysis System) for measuring acoustic wave velocities, resistivity, and some mechanic variables of hydrate-bearing samples (Riedel et al., 2010). Later the FLECAS system was modified into LARS (Large laboratory Reservoir Simulator) that uses a large pressure vessel made of steel, having an inner diameter of 600 mm and a depth of 1500 mm (Schicks et al., 2011). Multipurpose systems for measuring acoustic and other properties of natural and manmade samples that enclose gas hydrates were also created in Japan (Uchida et al., 2003) and China (Li et al., 2011).

Choosing the model of relations between hydrate contents and velocities of acoustic waves in sediments is another important task besides the laboratory measurements. This choice can stem from theoretical models of two main types, with averaging (Lee et al., 1996) or effective (Dvorkin et al., 1999) approximations, which however are controversial in application to real data (Hu et al., 2010). Laboratory experiments also show that acoustic velocities in hydrate-bearing sediments largely depend on their lithology, structure, and texture, as well as on the amount and form of hydrates and pore fluid type (Akselrod, 2009). Thus, gaining ever more experimental acoustic data on hydrate-bearing sediments is crucial for placing more rigorous constraints on these relationships and updating the respective theoretical models. Specifically, it is important to study the physical properties of freshwater hydrate-bearing sediments in the territory Russia, where methane hydrates were discovered in the sediments of Lake Baikal (Khlystov, 2006).

Laboratory acoustic measurements of hydrate-bearing samples had been impossible in Russia before we created the respective instrument (Duchkov et al., 2016) proceeding from the experience of studying other physical properties of such samples (Duchkov et al., 2006). The new system was used in numerous experiments on the formation of gas hydrates in pores of unconsolidated sediments (quartz sand) and subsequent measurements of travel times of acoustic waves. This paper provides a brief description of the system, the modeling methods, and preliminary analysis of the results.

Laboratory experimental setup

The system (Fig. 1) consists of a cylindrical steel pressure vessel, with a working pressure range to 45 MPa, an inner diameter of 30.6 mm, and 10–50 mm high, where samples (*4*) are placed during experiments. Axial and lateral pressure is transferred to the samples via anvils (*1*) and (*5*) and a cylindrical valve. Liquid and gas injected through special channels in anvils create the required pore pressure monitored by two sensors P_1 and P_2. Temperature is maintained by a thermostat and a cooling coat (*3*) and monitored by sensors T_1 and T_2 mounted into the coat and the upper anvil. Polar ultrasonic sensors made of piezoceramic discs 18 mm in diameter and 2 mm thick mounted on anvils (*1*) and (*5*) generate and receive *P* and *S* waves (the polarity of *S*-wave sensors is consistent in the lower and upper anvils). The sensors have a resonance frequency of 75 MHz (working frequency range 300–700 KHz, theoretical power limit 1.8 W) and are fixed tightly to the samples with lead protectors to attenuate parasitic noise. Axial pressure is applied to unconsolidated sediments (at least 10 MPa) to ensure good contact with the anvils. Acoustic waves are generated by a high-voltage pulse generator (minimum pulse duration 5 ns, frequency range from 0.1 to 50 MHz) and recorded by a 2-channel digital oscillograph, with a transmission band of 200 MHz and a sampling rate 500 MHz for both channels.

Our experiments on preparing hydrate-bearing samples and their acoustic measurements lasted from one to four days. The long-term measurements became possible due to a specially designed automatic operation, with connection to PC and data exchange (Fig. 2). The channels of *P* and *S* waves are switched according to PC signals with a switch based on the *Seeduino* microcontroller. PC also provides the control of the oscillograph (transfers triggering and recording parameters and oscillograph recordings) and the thermostat. The digital multichannel measuring unit MIT-8 records temperature and pressure data.

The user's graphic interface for the control of instruments and experiment runs was designed as part of the *LabView* package using the *PicoSDK 10.6.10* library (for the oscillograph) and the *NI VISA* library (for connection with other system units via the RS-232 interface, COM port).

The designed program can

(1) specify parameters of the experiment (duration, thermostat programming, ports, sampling rate, parameters of acoustic measurements, etc.);

(2) start the run;

(3) run cyclic measurements: two temperature channels (room temperature and temperature inside the upper anvil); four pressure channels (axial and lateral compression, pore pressure on the upper and lower anvil); axial strain and velocities of acoustic *P* and *S* waves;

(4) visualize data in real time (temperature and pressure curves and last *P* and *S* traces) and save them to disc (in ASCII format).

All control parameters, except information on connected ports, can change in the course of the experiment. Real-time