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GASES AND GAS HYDRATES IN THE EARTH'S CRYOSPHERE

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INFLUENCE OF GAS COMPOSITION AND PRESSURE ON THERMOPHYSICAL PROPERTIES OF GAS-SATURATED FROZEN AND THAWED SANDS

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Results of the experimental study of the gas composition and pressure effects on thermal conductivity and heat capacity of frozen and thawed sand saturated with different gases (nitrogen, methane, carbon dioxide, and a mixture of gases (50% CH_4 + 50% CO_2)) have been presented. According to results, the maximum gas pressure was set below the pressure of the gas hydrate formation. The experiments were conducted in a specially designed pressure chamber, which allowed to measure thermophysical characteristics of the gas-saturated sediments at temperatures below and above freezing point. The first ever data of gas pressure effect on thermal conductivity and heat capacity in the freezing sand saturated with different gases were obtained. The results revealed that thermophysical characteristics of the unfrozen gas-saturated sandy samples are practically independent of the gas composition and pressure. It has been established that a pressure increase in several saturating gases (N_2, CH_4, a CH_4 + CO_2 mixture, and CO_2) cause a reduction of thermal conductivity of the frozen sand, against an increase in the heat capacity. Carbon dioxide as highly soluble gas affecting the composition of unfrozen pore water, has the greatest influence on changes in the thermophysical parameters.

Gas-saturated sediment, freezing under gas pressure, experimental modeling, thermal conductivity, heat capacity, methane, carbon dioxide, nitrogen

INTRODUCTION

Thermophysical properties are important characteristics of freezing and thawing soils. Modeling of thermal processes during freezing of gas-saturated talik zones is impossible without the knowledge about variations of thermophysical parameters of gas-bearing sediments. It is a common fact that long-term freezing of gas-saturated dispersed sediments may lead to cryogenic concentration (expulsion of gas component by the freezing front) of fluids, and affiliated pore pressure excess in the thawed gas-saturated zone [*Chuvilin et al., 2000; Yakushev, 2009; Kraev et al., 2017; Chuvilin, Davletshina, 2018*]. Gas pressure is also contributed by freezing of gas-saturated closed sublake taliks (zones of unfrozen ground beneath lakes) in permafrost. In this case, pressure buildup processes can develop, which translates to a significant increase in pore pressure in the freezing zone leading thereby to ground deformations on the surface, frost-heaving and frost fracturing [*Gevorkyan, Koreisha, 1993; Grechishchev et al., 2012*], including events of cryovolcanism (accompanied by debris scattering and caving-in processes over distances measuring several tens of meters from the exploded structure) in the overlying permafrost horizon, and even to formation of gas blowout craters [*Bogoyavlenskiy, 2014; Bogoyavlenskiy, Garagash, 2015; Kizyakov et al., 2015; Khimenkov et al., 2017; Leibman et al., 2017; Buldovicz et al., 2018*]. The latter can be viewed as a source of potential risk to the nearby engineering constructions. In this regard, providing recommendations for prediction of the occurrence and timely detection of such "explosion hazardous" gas-saturated geological structures in the areas of economic development of the northern West Siberia is a high-priority task of practical importance. Nowadays, however, this problem solution would be impossible without modeling thermal and geomechanical processes in a confined gas-and-water-saturated zone exposed to freezing with due account of the influence of increasing pore pressure on the phase composition of pore-filling substance, as well as on thermal and mechanical characteristics of the investigated sediments.

Despite the relevance and applied significance, the thermal characteristics of gas-bearing rocks, especially those exposed to pressure, have thus far been largely understudied. Regarding the freezing sediment, most of the data available relate to the influence of the degree of pores filling with water or ice under pressures close to atmospheric [*Ershov et al., 1987; Komarov, 2003; Cheverev, 2004*]. More recent data provided in the literature include thermal conductivity estimates for gas-saturated bottom sediments in the Arctic seas shelf [*Chuvilin et al., 2013, 2015*], and for gas-saturated rocks in the unfrozen state depending on the gas component composition [*Chuvilin, Bukhanov, 2017*].

The available experimental data are largely insufficient for evaluation of the influence of the component composition and pressure of gas on the thermophysical characteristics of gas-saturated sediments during freezing, considering changes in the amount of liquid phase in gas-saturated soils with increasing pressure [*Istomin et al., 2018*]. The proposed problem formulation for the conducted experimental studies of thermophysical characteristics of gas-saturated dispersed sediments freezing under pressure is novel both in Russia and abroad.

EXPERIMENTAL METHODOLOGY

The thermophysical characteristics of a gas-saturated sediment with gas exposed to pressure were determined using a specially designed experimental system (Fig. 1) consisting of a thermal (climatic) chamber with working volume 0.5 m^3 which permitted to preset the required temperature; an original pressure cell equipped with gas injection system and data recording system for temperature and pressure control in the soil sample, and a special gauge for measuring thermal conductivity and heat capacity. The data on changes in thermobaric conditions in the pressure cell with the sample were recorded using an analog-to-digital converter (ADC) coupled with a computer.

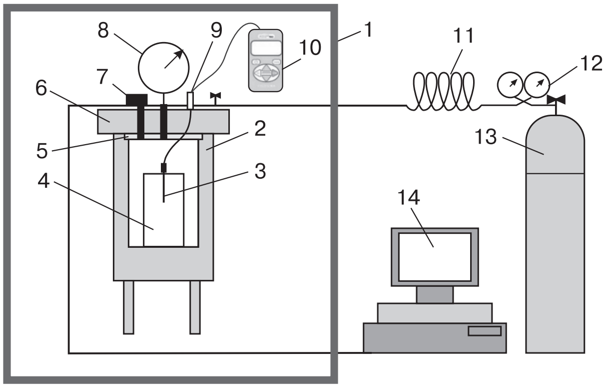

Fig. 1. Schematic of the experimental setup.

1 – climate-control chamber; 2 – pressure cell; 3 – sensor for thermal conductivity and heat capacity; 4 – container with the sample; 5 – tephlon gaskets; 6 – steel lid; 7 – pressure gauge; 8 – digital pressure gauge (manometer); 9 – thermal conductivity sensor port; 10 – KD-2 Pro tool; 11 – gas tube; 12 – pressure regulator; 13 – gas bomb; 14 – PC with ADC.

A pressure cell (100 mm high and 91 mm in diameter) with a working volume of about 0.7 L was filled by a container (100 mm high and 45 mm in diameter) with a water-saturated sample. The dual needle SH-1 sensor (length and diameter: 30 × 1.3 mm) of the KD-2 Pro tool (thermal properties analyzer) placed inside the sample permitted to measure thermophysical characteristics – thermal conductivity and volumetric heat capacity – with the precision to 10 %; one measurement was taken during ca. 2 minutes. The previous experience of using the KD-2 Pro tool for the study of frozen, thawed and hydrate-containing sediments at atmospheric pressure is described in the works authored by E.M. Chuvilin and B.A. Bukhanov [*Chuvilin, Bukhanov, 2013, 2019; Chuvilin et al., 2015*].

The samples chosen as the object of the experimental study were deformed natural sand of glacial-marine origin (gmQ_{II}^{2-4}), collected while drilling parametric wells within the Kharasavey oil and gas condensate field. The constituent minerals were identified by X-Ray diffractometry. The particle size distribution of fine and medium-grained sand (classification after E.M. Sergeev) were determined according to GOST 12536-2014 [*State Standard, 2014*]:

Particle size distribution, %	1.7	44.5	40.5	5.2	4.1	1.0	1.0	2.0
Particle diameter, mm	1–0.5	0.5–0.25	0.25–0.1	0.1–0.05	0.05–0.01	0.01–0.005	0.005–0.001	<0.001

The physical characteristics of the test sample determined following the standard procedures prescribed in GOST 5180-2015 [*State Standard, 2015*] and SNiP 2.02.04-88 [*Building Code, 1990*] included: moisture content 16 %, density 2.05 g/cm^3, porosity 0.34, pore filling degree 0.83, specific surface area 0.3 m^2/g. Sand consists dominantly of quartz (94 %), and also contains kaolinite and chlorite (3 %), microcline (2 %), illite (0.5 %) and minor traces of amphibole. The salinity estimated by chemical analysis of aqueous extract (supernatant) is 0.07 %.

The major-ion chemistry of supernatants (mg-eq./100 g) was: HCO_3^- 0.09, Cl^- 0.27, SO_4^{2-} 0.12, Ca^{2+} 0.09, $Na^+ + K^+$ 0.35, Mg^{2+} 0.04; dry residue (total salts) 67.5 mg; pH 7.1.

The method for experimental determination of thermal conductivity and heat capacity of the investigated pressurized gas-saturated sediment involved preparation of a soil sample with prespecified water content. The required (initial) moisture content was achieved by mixing air-dry soil with distilled water. The output mass was left for 30 min at room temperature, to achieve uniform distribution of water. Then, the prepared wet soil (sample) was compacted layer-by-layer in a cylindrical container [*Chuvilin, Guryeva, 2009*].

The prepared soil sample was placed into the pressure cell which was tightly sealed and vacuumed, and then the pressure cell with the sample was filled with precpecified hydrate-forming gas to pressure below hydrate formation. The pressure cell was relocated into the thermal chamber.

At the beginning of the experiment, the thermophysical characteristics of the soil sample were measured under atmospheric pressure first at positive and then negative (−6 °C) temperatures. After that, cold gas (−6 °C) was injected into the pressure cell to the maximum pressure (specific for each gas) and then the pressure cell was exposed to cooling–heating in a −6...+10 °C cycle, with the thermal conductivity and heat capacity measured before and after the freezing. Then, the gas pressure in the pressure cell was reduced stepwise at a constant temperature (+10 °C), while the cooling-heating cycle was repeated at each step, with concomitant measurements of the thermophysical characteristics.

The hydrate-forming gases involved in the experiments are given in the order as follows: nitrogen N_2, methane CH_4; methane – carbon dioxide mixture ($CH_4 + CO_2$) in a ratio (%) 50:50, and carbon dioxide CO_2. Before applying a new type of hydrate-forming gas, the pressure cell containing the sample was vacuumed. The pressure was set depending on the type of gas within the prespecified ranges as follows: 0.1–4 MPa for N_2, 0.1–2 MPa for CH_4, 0.1–0.8 MPa for CO_2, and 0.1–1.5 MPa for a 50% CH_4 + 50% CO_2 mixture. In all the cases, gas pressures in the test cell were set below the equilibrium value of hydrate formation.

EXPERIMENTAL RESULTS AND DISCUSSION

Results of the experimental study of gas pressure influence on the thermal conductivity and heat capacity of gas-saturated sandy soil are presented in Fig. 2 and in Table 1.

The obtained experimental estimates for thermophysical properties of a sandy sample saturated with different gases show that while the thermal conductivity of wet sand sample naturally increases after freezing (Fig. 2, *a*, *c*), its specific heat capacity decreases (Fig. 2, *b*, *d*). The influence of gas pressure and gas type on the thermophysical properties of the sand sample is most clearly manifest in the frozen state of the sample (Table 1).

In the case of nitrogen saturation of the sand sample (water content *W* = 16 %), its conversion from thawed to frozen state (−6 °C) was characterized by a 60–70 % increase in the thermal conductivity and a 15–20 % decrease in the heat capacity. At this, when the N_2 pressure increased to 4 MPa in the frozen sample, its thermal conductivity showed a slightly (2–3 %) decreasing trend. The coefficient of thermal conductivity reduction λ induced by gas pressure was defined as the ratio of thermal conduc-

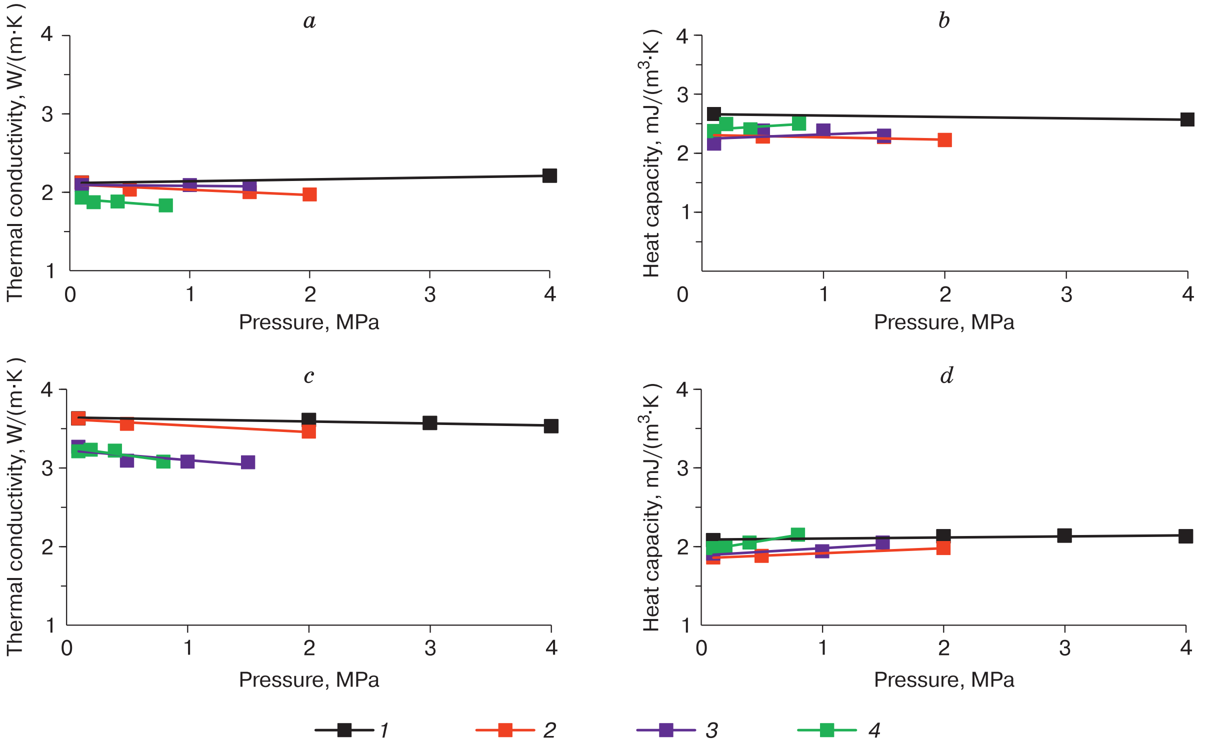

Fig. 2. Variations of thermal conductivity and heat capacity of gas-saturated sand (*W* = 16 %) for the thawed (*a*, *b*) and frozen (*c*, *d*) samples depending on gas pressure.

Gas composition: *1* – N_2; *2* – CH_4; *3* – $CH_4 + CO_2$; *4* – CO_2.

Table 1. **The influence of gas composition and pressure on thermal conductivity (λ) and heat capacity (*C*) of gas-saturated sand (*W* = 16 %) before and after freezing**

Gas type	Pressure, MPa	Before freezing		After freezing	
		λ, W/(m·K)	*C*, mJ/(m^3·K)	λ, W/(m·K)	*C*, mJ/(m^3·K)
N_2	0.1	2.12	2.66	3.63	2.08
	2.0	–	–	3.61	2.13
	3.0	–	–	3.57	2.14
	4.0	2.21	2.57	3.53	2.13
CH_4	0.1	2.12	2.31	3.63	1.86
	0.5	2.03	2.28	3.56	1.88
	1.5	2.00	2.27	–	–
	2.0	1.97	2.22	3.46	1.98
$CH_4 + CO_2$	0.1	2.09	2.16	3.27	1.91
	0.5	–	2.38	3.09	–
	1.0	2.09	2.38	3.08	1.94
	1.5	2.07	2.29	3.07	2.05
CO_2	0.1	1.93	2.37	3.21	1.98
	0.2	1.87	2.49	3.23	1.99
	0.4	1.88	2.40	3.22	2.05
	0.8	1.83	2.50	3.08	2.15

tivity to the prespecified gas pressure range. In the gas-saturated frozen sample (*W* = 16 %), the reduction coefficient λ equals [(W/(m·K))/MPa]: 0.03 for N_2, 0.08 for CH_4, 0.13 for a CH_4 + CO_2 mixture, 0.2 for CO_2.

The heat capacity of the nitrogen-saturated frozen sand sample tends to slightly increase as the applied pressure increases up to 4 MPa (about 0.02 mJ/(m^3·K) per 1 MPa).

The thermal conductivity of the frozen sand saturated with methane shows a 2–5 % decrease when the gas pressure increases to 2 MPa. The coefficient of pressure-induced reduction of thermal conductivity in a CH_4-saturated sample was 0.08 W/(m·K)/MPa. The heat capacity in this frozen sand sample exposed to gas pressure increased up to 2 MPa changes slightly (≤6 %), and the increase factor for heat capacity equals to 0.05 (mJ/(m^3·K))/MPa.

A 45–55 % increase was reported for the thermal conductivity of sand saturated with a CH_4 + CO_2 mixture (50/50) during its freezing. With the increased pressure of this gas mixture (from 0.1 to 1.5 MPa), the thermal conductivity of frozen sample shows an explicable 6–7 % decrease, while the reduction coefficient of thermal conductivity was 0.13 (W/(m·K))/MPa. At this, an increase in the heat capacity of sand freezing under pressure within the specified range reaches 7 % (around 0.08 mJ/(m^3·K) per 1 MPa).

The case of pure CO_2 saturation of the sand sample was marked by a 65–75 % increase in the thermal conductivity of the sand sample during its freezing. Within the studied pressure range (0.1–0.8 MPa), the coefficient of thermal conductivity reduction is 0.2 (W/(m·K))/MPa, the increase factor for heat capacity is 0.26 (mJ/(m^3·K))/MPa. As such, the tendencies are dictated primarily by the decreased freezing point and increased content of unfrozen water in the frozen sediments as the carbon dioxide pressure has risen [*Istomin et al., 2018; Chuvilin et al., 2019*].

Analysis of the experimental data demonstrates that the thermal conductivity of the sand saturated with different gases under pressure is explicably higher in the frozen, than in thawed sand. This is largely accounted for a significant differentiation of the thermal conductivity of water in the liquid and solid states.

Both thermal conductivity and heat capacity of the frozen sand sample saturated with gases are found to be more strongly dependent on the gas pressure and chemical composition, as compared to the thawed sample. The influence of gas pressure on the decrease in thermal conductivity and increase in heat capacity of the gas-saturated frozen sample grows in the following order: N_2, CH_4, CH_4 + CO_2, CO_2. At this, variation of the coefficient of pressure-induced thermal conductivity reduction represents an order of magnitude change, from 0.03 to 0.2 (W/(m·K))/MPa. The material effect of CO_2 on the thermophysical properties of frozen sand is associated with this gas ranked as highly soluble in pore water [*Istomin et al., 2018*] and as capable to inflate unfrozen water content in the frozen soil as gas pressure increases. In addition, the thermophysical properties of the pressurized gas-saturated freezing sand are affected by other factors, such as the thermophysical properties of gases [*Vargaftik, 1972; Vargaftik et al., 1990; Babichev et al., 1991*]. Thus, the data provided by A.P. Babichev and colleagues [*1991*] indicate that thermal conductivity in the considered group of gases comprising CH_4, N_2, CO_2 decreases to 0.0342, 0.0257 and 0.016 W/(m·K), respectively, at 0.1 MPa and under room temperature. It was previously noted that with the gas pressure increasing to 4 MPa, thermal conductivities of the experimental gases tend to show a minor increase [*Vargaftik, 1972*]. The effect of structural and textural characteristics of the frozen sample related to the pore water crystallization behavior during freezing, should not be neglected either. The thermophysical properties of gas-saturated soil at negative temperatures will be most strongly influenced by the pressure-dependent content of unfrozen water, the negative temperature value and the type of gas. This however requires more in-depth research.

CONCLUSIONS

A method for determining the thermophysical parameters of gas-saturated soils under pressure below hydrate formation at positive and negative temperatures was developed during the experimental study. The obtained results provided new data and insights into the gas pressure effects on the thermal conductivity and heat capacity of the frozen and thawed sandy samples saturated with nitrogen, methane, a mixture of gases (50% CH_4 + 50% CO_2), or carbon dioxide acting as hydrate forming gases.

It is shown that in the case of the thawed sample saturated with a hydrate-forming gas, thermal conductivity and heat capacity of sand depend little on the gas composition nor pressure. Alternatively, during the gas-saturated sand sample freezing under pressure, its thermal conductivity shows a 35–70 % increase, against the backdrop of a 10–20 % decrease in the specific heat capacity.

It was found that the thermophysical characteristics of a frozen gas-saturated sandy sample are largely controlled by the gas pressure and its chemical composition: the thermal conductivity of the gas-saturated frozen sample decreases, while its heat capacity increases as the gas pressure exceeded the equilibrium pressure for the entire sequence of the gases involved in the experiment (N_2, CH_4, a mixture of gases (CH_4 + CO_2) and CO_2). The obtained estimates for their pressure-induced variations are: from 0.03 to 0.2 W/(m·K) per 1 MPa for the coefficient of thermal conductivity reduction, and from 0.02 to 0.26 mJ/(m^3·K) for the heat capacity increase factor.

The experiment results demonstrated that, when injected into soil sample, CO_2 gas entails a decrease in the thermal conductivity of frozen sand, which is at least 10 % lower, than thermal conductivities obtained for sand samples saturated with nitrogen and methane. This effect is associated with a decrease in the freezing point of pore water in pressurized gas-saturated soil and inflated content of the unfrozen water in frozen (ice-bearing) soils as the CO_2 pressure increases.

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