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Effect of gas hydrates on physical properties of permafrost

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Abstract. Permafrost stores large amounts of natural gas in free and hydrate (clathrate) forms. Intrapermafrost gas hydrates were revealed in frozen core samples recovered from test, exploration, and production wells in the Arctic oil and gas fields. Being similar to ice in many respects, gas hydrates can be mute for geophysical surveys. Meanwhile, laboratory experiments show that the presence of a gas hydrate component in pore moisture affects considerably some parameters of frozen sediments, as well as processes in evolving permafrost. Hydrate-bearing permafrost has a lower thermal conductivity and permeability but a higher geomechanic strength than hydrate-free frozen ground. Gas hydrates also influence some geophysical variables, such as electrical resistivity which becomes higher at higher hydrate contents.

Accumulation of gas hydrates changes the relative percentages of pore moisture components (ice-hydrate-liquid water) in systems comprising ice, free gas, and gas hydrates, which has bearing on the physical properties of permafrost. The presence of gas hydrates increases the strength and resistivity of permafrost and, on the other hand, tends to reduce its permeability and the rates of heat and mass transfer.

Key words: permafrost, gas hydrates, pore ice, permeability, resistivity, methane

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Introduction

Natural gas hydrates (primarily methane hydrates), which store at least 170 m³ of methane in each cubic meter of hydrate at relatively shallow depths, have a high potential as an unconventional resource of hydrocarbon gases comparable to the total of the conventional resources (Yakushev et al., 2014; Chong et al., 2016; Varenichev et al., 2022).

In this respect, gas hydrate research over the past several decades has focused on improving technologies for methane recovery from natural hydrate reservoirs, but mostly in subsea settings where around 95% of known gas hydrate accumulations presumably reside (Wei et al., 2024; Max, Johnson, 2016; Yamamoto

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et al., 2022). The remaining 5% of gas hydrate occurrences are associated with permafrost, on land or on the Arctic shelf (Chersky et al., 1973; Makogon, 1985; Istomin, Yakushev, 1992; Max, 2000; Yakushev, 2009; Ruppel, 2015; Matveeva et al., 2024). The stability of gas hydrates in high-latitude environments is controlled by the properties and evolution of permafrost. Gas hydrates can exist beneath permafrost at low positive temperatures or within permafrost at subzero temperatures. Subpermafrost gas hydrates were reported from the Mackenzie Delta, the Alaska North Slope (Mount Elbert, Northwest Eileen, and Ignik Sikumi sites), the Messoyakha gas field, and Tibet (Agalakov, 1997; Ginsburg, Novozhilov, 1997; Dallimore et al., 1999; Collett et al., 2011; Makogon, Omelchenko, 2012; Li et al., 2017; Boswell et al., 2022). Intrapermafrost gas hydrates were retrieved from core samples of the Taglu 92 GSC well in the Mackenzie Delta, Canadian Actic coast (Dallimore, Collett, 1995) and in the Olenyok River mouth, Central Siberian Arctic coast (Chersky

et al., 1973). Furthermore, there is a wealth of implicit evidence for possible existence of gas hydrates in the Arctic shelf and northern West Siberian oil and gas fields (Chuvilin et al., 2000; Yakushev, Chuvilin, 2000; Yakushev, 2009; Chuvilin et al., 2020).

Gas hydrates can form in permafrost by several mechanisms: perennial freezing of subpermafrost gas reservoirs; hydration during upward migration (cryogenic expulsion) of deep gas which reaches a P-T zone of hydrate stability (Yakushev, 2009); loading from ice sheets which expand the hydrate stability zone (Trofimuk et al., 1986).

Hydrate formation in gas-saturated freezing and frozen soils was simulated in laboratory experiments (Chuvilin, Guryeva, 2009; Chuvilin, Davletshina, 2018; Davletshina, Chuvilin, 2018; Chuvilin et al., 2019).

Although intrapermafrost gas hydrates were discovered over fifty years ago (Chersky et al., 1973), they remain quite poorly studied, mainly because they are hard to discriminate from ice in geophysical records (especially, in seismic data) according to density, acoustic velocity, and other parameters. Nonetheless, it is known from limited literature that the presence of gas hydrates in permafrost interferes with its temperature, mechanical strength, permeability, and resistivity patterns.

Thus, understanding the effect of gas hydrates on the basic permafrost properties can contribute to the knowledge on ice- and hydrate-bearing compounds in porous media. Moreover, this knowledge has practical implications for wellbore stability in Arctic subpermafrost gas reservoirs, which is controlled by thermal and mechanical interactions of wells with the drilled ice- and hydrate-bearing permafrost.

Methods

The effect of the gas hydrate component present in the pore moisture on the properties of freezing and frozen soils was studied in laboratory experiments simulating hydrate formation on specially designed high-pressure systems. The experiments were applied to natural sand and silt sampled in gas fields of northern West Siberia and the Arctic shelf, as well as laboratorymade sand samples and sand-silt-clay mixtures.

The inventory of instruments included previously described systems (Chuvilin, Bukhanov, 2014, 2017; Chuvilin, Grebenkin, 2015; Yang et al., 2019) for measuring thermal, geomechanical, flow, and geophysical parameters (Table 1) at varied relative percentages of hydrate, ice, liquid water, and gas phases in the pore moisture.

Frozen hydrate-bearing samples with high hydrate contents ($\geq 50 \text{ vol.\%}$) were prepared for experiments under above-equilibrium pressures of the hydrateforming gas, as in (Chuvilin, Davletshina, 2018), using 99.998% methane or 99.995% carbon dioxide in gas bombs pressurized to 10.0 and 6.0 MPa, respectively. Hydrate saturation (S_{ι}) and hydrate coefficient (K_{ι}) , the main parameters of gas hydrates, were estimated by the PVT method from sample temperatures and gas pressure data (Chuvilin, Davletshina, 2018; Chuvilin et al., 2019).

Hydrate saturation $(S_h, \%)$ refers to percentage of pore space occupied with gas hydrate and is estimated from volumetric content of hydrate H_{α} (%) and porosity n (u.f.):

$$S_h = \frac{H_v}{n}. (1)$$

Hydrate coefficient $(K_h, u.f.)$ refers to percentage of pore ice converted to hydrate and is given by

$$K_h = \frac{W_h}{W},\tag{2}$$

where W_{h} is the dry weight percentage of pore moisture converted to hydrate and W is the initial moisture content (wt. %). W_h was estimated from the weight of pore hydrate M_h which, in turn, was calculated with reference to gas consumption assuming the hydrate formulas CH₄ · 5.9 H₂O for methane and CO₂ · 6.1H₂O for carbon dioxide.

The thermal conductivity λ (W/(m·K)) of frozen hydrate-bearing samples was estimated at a gas pressure above the equilibrium using a gas hydrate thermal testing system and a METER Group KD2 Pro thermal analyzer (USA). The gas hydrate system includes a built-in unit for thermal conductivity measurements on the steady-state basis, which was originally designed by our team in collaboration with the Institute of Nature Management (Minsk, Belarus) (Brovka, Romanenko, 2010). The KD2 Pro thermal analyzer is equipped with an SH-1 dual-needle thermal diffusivity and specific heat sensor (Chuvilin et al., 2023). The samples tested on the gas hydrate system under the steady-state thermal regime were 100 m high and 10 mm thick hollow cylinders placed between two coaxial cylinders. The measurements with the KD2 Pro analyzer were applied to ice- and hydrate-bearing cylindrical samples, 90 mm high and ~60–70 mm in diameter. In both cases, samples were overheated to no more than 0.5 °C. The thermal conductivity estimates were accurate to \pm 5%, as in our previous studies (Chuvilin, Bukhanov, 2019).

The geomechanical strength (σ , MPa) of frozen hydrate-bearing samples with different relative percentages of pore ice and hydrate was measured on a VJ Tech *Tri-Scan 250* triaxial testing system (UK) as part of a collaborative project with Heriot-Watt University (Edinburgh). Strength was expressed via

Properties	Estimated variables	Instruments	Reference
Thermal	Thermal conductivity (W/(m·K)	Gas hydrate system, with a built-in unit for thermal conductivity measurements on the steady-state basis designed at Skoltech (Russia) in collaboration with the Institute of Nature Management (Minsk, Belarus) METER Group <i>KD2 Pro</i> thermal analyzer (USA) with an SH-1 dual-needle sensor (USA)	Chuvilin and Bukhanov, 2014; Chuvilin and Bukhanov, 2017; Chuvilin et al., 2023
Geomechanical	Shear strength (MPa)	VJ Tech <i>Tri-Scan 250</i> triaxial testing system (UK) designed for gas hydrate research	Yang et al., 2019
Flow	Effective gas permeability (mD)	EcogeosProm flow testing system (Russia)	Chuvilin and Grebenkin, 2015; Chuvilin et al., 2021
Geophysical	Resistivity (kOhm·m)	Wenner four-point array for resistivity measurements in hydrate-bearing soil	ASTM G57-20

Table 1. Main physical properties of hydrate-bearing rocks relevant to this study

the deviatoric stress ($\sigma_1 - \sigma_2 = \sigma_3$), i.e., the difference between the total stress (axial pressure) and hydrostatic stress (constant confining pressure). The Tri-Scan 250 system was designed for studies of shearing strength and deformation behavior in gas-saturated hydrate-bearing rocks under gas pressure (Yang et al., 2019). The samples were made in laboratory as 100 m high cylinders, 50 mm in diameter, composed of fine silica sand mixed with silt and bentonite clay (montmorillonite). The samples were frozen, saturated with hydrate, and exposed to aboveequilibrium methane pressure ~5 MPa (conditions of gas hydrate stability) and temperatures of -3 °C and −10 °C. The samples were sheared at a constant rate of 0.1 mm/min.

The gas permeability (K, mD) of ice- and hydratebearing samples at different pressures and temperatures was studied on EcogeosProm flow testing system (Russia) which can operate at large P-T ranges of -15 to +30°C and up to 10 MPa (Chuvilin, Grebenkin, 2015). Effective gas permeability $(K_{\rm rel}, u.f.)$ was estimated from pulse decay as the $K_{\rm hs}/K_{\rm initial}$ ratio, where $K_{\rm hs}$ and K_{initial} are, respectively, effective gas permeability values for hydrate-saturated and initial hydrate-free frozen sample, ~30 mm in diameter and 30-40 mm high. The temperature was maintained to an accuracy of ± 0.5 °C using a Binder MK240 climate chamber (USA).

The geophysical responses were studied under a gas pressure of 4-5 MPa in a *Top Industrie* high-pressure cell (France). Electrical resistivity (ρ , kOhm·m) was measured by a dc Wenner four-point array of equally spaced brass rods, 3.5 mm in diameter (Grebenkin et al., 2024), in cylindrical samples ~80 mm high and 38 mm in diameter. The temperature was maintained with a Julabo FP40 liquid cryostat to a precision of \pm 0.2 °C.

Results: changes of physical properties in response to hydrate formation

The presence of gas hydrates in the pore space of sediments affects considerably their physical properties which differ from those of other pore moisture components (ice, liquid water, and gas), such as geomechanical strength, thermal conductivity, permeability, and resistivity. The physical properties of hydrate-bearing permafrost are also sensitive to temperature and pressure variations. This study focuses on the effect of pore gas hydrates on the properties of frozen samples at the P-T conditions of pore hydrate stability: above-equilibrium gas pressures and negative temperatures.

Thermal conductivity

Ice and gas hydrates differ markedly in thermal conductivity, which is thus sensitive to the presence of gas hydrates in the pore space of frozen sediments. The experiments revealed a notable decrease (~ 22%) in thermal conductivity upon transition from pore ice to gas hydrate (Figure 1), from 2.32 W/(m·K) at $K_{\rm h}$ = 0 to 1.80 W/(m·K) at $K_h = 0.5$ in fine sand (W = 19%). However, further hydrate formation $(K_b > 0.5)$, when the thermal conductivity became more controlled by the

soil skeleton, caused less prominent changes (Chuvilin, Bukhanov, 2017). In general, the thermal conductivity decrease in frozen sediments associated with pore ice-to-hydrate conversion results from the loss of the more conductive ice component and the gain of the hydrate component, with its thermal conductivity almost four times as low: $2.22-2.35~\text{W/(m\cdot K)}$ against $\sim 0.6~\text{W/(m\cdot K)}$. Correspondingly, the thermal conductivity of frozen silt and silty sand reservoirs can be expected to become 15-20% lower on average as up to 40-50~vol.% of pore ice converts to methane hydrate (Chuvilin, Bukhanov, 2017; Chuvilin et al., 2023).

Shear strength

The tests constrained the relationships of shear strength with hydrate saturation (S_h , %) and hydrate coefficient (K_h , u.f.) in ice- and hydrate-bearing samples (W = 15%, $\rho = 2.05$ g/cm³, $\varphi = 0.34$) at negative temperatures of -3 °C and -10 °C (Figure 2). The total saturation (ice+hydrate) in the tested samples approached 75–80%.

Shear strength correlates linearly with hydrate saturation, at both warmer (-3 °C) and colder

(-10 °C) negative temperatures (Figure 2a), i.e. the geomechanical properties of frozen sediments are sensitive to the presence of a hydrate component in the pore moisture as pure hydrate has a higher shear strength than ice (Stern et al., 1996). The strength can increase further upon cooling, at the same hydrate coefficient: from 9.6 MPa at -3 °C to 14.1 MPa at -10 °C (Figure 2b). This considerable change (~50%) may be due to a decreasing amount of liquid pore water: cooling from -3 to -10 °C was shown by recent NMR

relaxation experiments to cause a 25% decrease in the content of residual unfrozen water (from 1.2 to 0.9%) in frozen hydrate-bearing fine sediments, at a methane pressure of 5 MPa (Bukhanov et al., 2022). Therefore, the content of unfrozen pore water is another strength control in hydrate-bearing permafrost.

Gas permeability

Partial conversion of pore ice to hydrate leads to changes in the permeability of gas-bearing frozen sediments. Experiments with samples of 43% to 63% ice saturation revealed permeability decrease in response to hydrate formation (Chuvilin et al., 2021). Judging by the permeability ($K_{\rm rel}$) vs. hydrate coefficient ($K_{\rm h}$) patterns (Figure 3), the effect of pore hydrate formation on gas permeability is stronger at higher initial ice saturation $S_{\rm h}$.

The conversion of pore ice into gas hydrate increases the total (ice+hydrate) saturation due to about 15% difference in specific volumes of the two pore moisture components, whereby the volume of voids, which are paths for gas flow, reduces correspondingly. At an initial ice saturation of \sim 63%, even minor change in the volumetric percentage of voids leads to permeability decrease larger than in the case of lower saturation (43%). Thus, the gas permeability of sediments with \geq 60% ice saturation can become tens of times lower upon pore hydrate formation whereas the respective decrease in less ice-saturated rocks (about 40%) hardly reaches a factor of two.

The variations of relative gas permeability in sediments with different ice saturation values are additionally controlled by the gas type (CH_4 or CO_2), at a negative temperature of -5 °C (Figure 4).

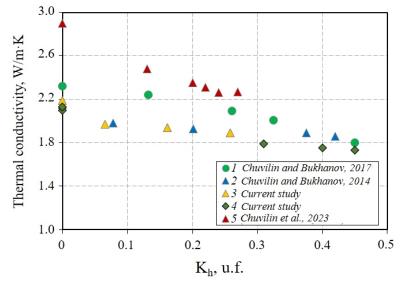


Figure 1. Thermal conductivity (λ) variations in sand and silty sand as a function of hydrate coefficient (K_h), at -5 °C. 1 – fine sand (quarry, vicinity of Lyubertsy, Moscow region), W = 19%, $\varphi = 0.40$; 2 – silty sand (Laptev Sea shelf, vicinity of the Tiksi site), W = 15%, $\varphi = 0.38$; 3 – silty sand (Yamburg gas field), W = 16%, $\varphi = 0.40$; 4 – silt (quarry, vicinity of Vorkuta), W = 16%, $\varphi = 0.40$; 5 – fine sand (South Tambey gas field), W = 16%, $\varphi = 0.37$

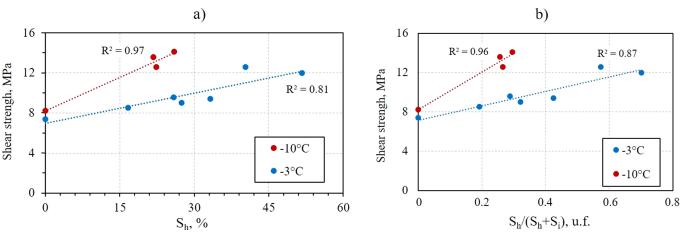


Figure 2. Shear strength of ice- and hydrate-bearing sand and silty sand as a function of hydrate saturation S_h (a) and hydrate coefficient K_h (b), at -5 °C

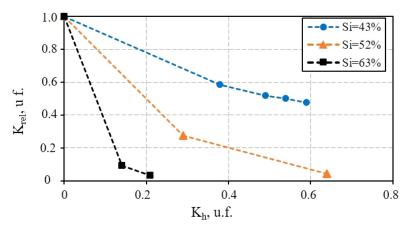


Figure 3. Relative gas permeability (K_{rel}) of frozen sand with ice saturation (S_i) of 43%, 52%, and 63% as a function of methane hydrate coefficient (K_h) , at -5 °C

The permeability of sand samples with initial porosity of 0.35-0.36 u.f., ice saturation of 49–54%, and $\sim 25\%$ CO₂ hydrate saturation was as low as 0.7 mD. The respective values for pore methane hydrate in sand samples with an initial porosity of 0.46-0.48 u.f. were 31-34% ice saturation, ~ 17% hydrate saturation, and 0.5–0.6 mD gas permeability (Figure 4).

The samples showed a quasi-linear correlation between gas permeability and volumetric percentage of voids (Figure 5).

Resistivity

The effect of pore gas hydrate on electrical resistivity was estimated as a function of hydrate coefficient in two runs (Figure 6).

The samples consisted of silica sand with 0.1–0.5 mm particle sizes (run RS1) and natural fine sand (run RS2), with initial parameters of 12–13% water content, ~ 0.37 u.f. porosity, 64% ice saturation, and resistivity values of 19.77 kOhm·m (RS1) and 28.81 kOhm·m (RS2). Hydrate formation was provided by injection of methane (run RS1) and carbon dioxide (run RS 2).

The experiment focused on resistivity responses to the formation of methane (run RS1) and carbon dioxide (run RS2) hydrates, at a constant negative temperature of -6 °C (Figure 6). Saturation with methane to a gas pressure of 6 MPa led to a moderate resistivity increase of < 10%, up to 21.72 kOhm·m, but the injection of CO, till a gas pressure of 2.9 MPa caused a more than three-fold resistivity decrease, down to 8.8 kOhm·m (Figure 6). The decrease in the latter case was due to a larger amount of unfrozen pore water produced by pore ice melting upon active interaction with CO₂.

In run RS1, the resistivity of the frozen sample became 2 kOhm m higher after 2% of pore ice had converted to methane hydrate (Figure 6) and then showed an almost linear correlation with hydrate coefficient: it increased by a factor of 1.5 (up to 31.85 kOhm·m) as K_h reached 0.17 u.f. and then became two times higher (41 kOhm·m) as 27% of pore ice converted to methane hydrate.

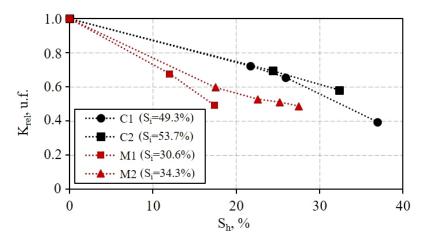


Figure 4. Gas permeability (K_{rel}) of frozen samples with different ice saturation percentages as a function of hydrate saturation with CO_2 (black) and CH_4 (red) hydrates (S_h)

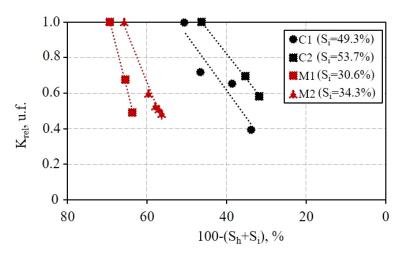


Figure 5. Relative gas permeability (K_{rel}) as a function of void volume percentage $(100-(S_h+S_i))$ in frozen samples during formation of CO_2 (black) and CH_4 (red) hydrates

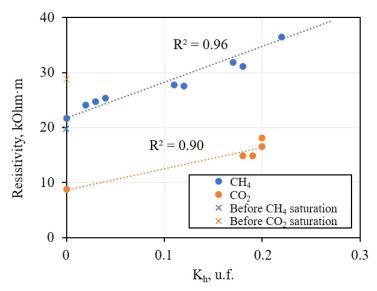


Figure 6. Resistivity of frozen sand samples (runs RS1 and RS2) as a function of hydrate coefficient, for hydrates of methane and carbon dioxide, at -6 °C

A similar pattern of K_h dependence of resistivity was obtained in run RS2, at a higher rate of pore ice-to-CO₂ hydrate conversion. The resistivity became 1.7 times higher (14.8 kOhm·m) at a hydrate coefficient of 18% and increased further to 18.1 kOhm·m as another 2% of pore ice converted to hydrate.

Thus, although the resistivity values differed in magnitude in the two runs, the relative increase in response to hydrate formation was similar and reached factors of 1.6 (methane hydrate) and 2 (CO_2 hydrate) at $K_b = 20\%$

Discussion

The new experimental results, as well as published evidence, indicate that the presence of pore gas hydrates causes a notable influence on the physical properties of permafrost. The main reason is that the considered variables (shear strength, thermal conductivity, gas permeability, and resistivity) have different values in ice and gas hydrates, but the relative percentages of pore moisture components (amount of liquid water) come into play as well in some cases. Therefore, the behavior of different variables controlled by hydrate content and above-equilibrium pressure may differ in both magnitude and trends of change (Table 2).

In general, the shear strength of permafrost is proportional to the percentage of the hydrate component ($K_h = 0.5$ corresponds to $\sim 50\%$ strength increase), because shear strength in gas hydrates is higher than in ice. On the other hand, gas hydrates differ from ice rheologically and are more prone to brittle failure while deformation in ice under a constant load is mostly ductile.

Thermal conductivity decreases in response to hydrate formation as gas hydrates are less thermally conductive than ice. The expected decrease for sand and silt is moderate: 15–20% at conversion of 50% of pore ice to hydrate.

Variations of gas permeability are controlled primarily by initial saturation. Conversion of pore ice to

hydrate increases total saturation as gas hydrates occupy a $\sim 15\%$ greater volume in the pore space, which reduces the volume of voids as gas flow channels. Therefore, frozen gas reservoirs with initial ice saturation as high as 60% or more become tens of times less permeable to gas upon hydrate formation, whereas the respective decrease at lower ice saturation ($\le 40\%$) is much smaller.

The phase transition from the less electrically resistive pore ice to more resistive gas hydrate expectedly leads to resistivity increase, which, on the other hand, may be also due to decreasing amount of residual liquid pore water. The content of unfrozen water in permafrost subject to hydrate formation tends to equilibrium (non-clathrate water), while the content of non-clathrate water is much lower than that of unfrozen pore water in hydrate-free frozen sediments and decreases with increasing gas pressure (Chuvilin, Istomin, 2012). Therefore, the resistivity of frozen hydrate-bearing sand can be expected to become 1.5 to 2 times higher as hydrate coefficient reaches 0.25. Note also that the reduction in the amount of unfrozen pore water in hydrate-accumulating sediments increases the geomechanical strength of permafrost.

The reported data generally confirm that the basic physical properties of permafrost containing pore ice, free gas, and gas hydrates are sensitive to the relative percentages of the pore moisture components (pore ice and gas hydrate), as well as to the amount of liquid pore water. Thus, the presence of pore gas hydrates increases the geomechanical strength of permafrost and, at the same time, reduces its capacity of heat and mass transfer.

Conclusions

Permafrost is a favorable environment for the formation and prolong existence of natural gas hydrates. Although being similar to ice in many respects, gas hydrates present in the pore space influence processes in permafrost associated with its evolution. The presence of a hydrate component in the pore moisture changes the thermal, electrical, geomechanical, and flow properties

Parameter	Hydrate coefficient	Trend	Magnitude of change
Shear strength	~0.5	+	~50%
Thermal conductivity	0.5	_	15-20%
Gas permeability	0.6	_	Several times to orders of magnitude
Resistivity	≤0.25	+	1.5-2 times

Table 2. Effect of hydrate formation on behavior of physical variables. Symbols "-" and "+" mean decrease and increase of values, respectively

of permafrost relative to hydrate-free sediments. It generally decreases the thermal conductivity and gas permeability of permafrost but increases its shear strength and electrical resistivity. In addition to the presence or absence of pore gas hydrates, the physical properties of permafrost are sensitive to pressure, temperature, and amount of residual liquid pore water.

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IN RUSSIAN

Влияние газогидратной компоненты на свойства мерзлых пород

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Одной из особенностей многолетнемерзлых пород является способность аккумулировать в себе крупные скопления природного газа, в том числе и в гидратной форме. Информация о наличии внутримерзлотных гидратосодержащих коллекторов была получена в результате исследования мерзлых кернов, поднятых в ходе параметрического бурения, а также на основе анализа многочисленных данных о газопроявлениях из интервалов многолетнемерзлых пород при бурении разведочных и добывающих скважин в Арктике. В настоящее время установлено, что газовые гидраты по ряду физических показателей очень сходны со льдом, поэтому их выявление в толще мерзлых пород при использовании стандартных геофизических методов (в первую очередь сейсмики) достаточно затруднительно. Однако, как показали результаты проведенных экспериментальных исследований, газогидратная компонента может оказывать существенное влияние на некоторые физические характеристики мерзлых пород и процессы, которые протекают в толщах мерзлых пород в ходе их существования и эволюции. Так, наличие поровых гидратов в мерзлых породах способствует с одной стороны снижению их теплопроводности и фильтрационных характеристик, а с другой - существенному повышению прочности льдосодержащего коллектора. Отдельно рассмотрено

влияние газогидратной компоненты на некоторые геофизические характеристики мерзлых грунтовых сред. В частности, отмечено закономерное повышение удельного электрического сопротивления мерзлых пород при увеличении гидратосодержания.

В ходе исследования показано, что изменения основных физических характеристик мерзлых пород в условиях накопления поровых газогидратов связаны не только с изменением соотношения поровый лед – поровый гидрат, но и с содержанием жидкой фазы воды в льдо-, газо- и газогидратных грунтовых системах. В итоге сделан вывод, что появление газогидратной компоненты с одной стороны существенно упрочняет мерзлый коллектор, а с другой проявляет тенденцию к снижению способности к тепло- и массопереносу.

Ключевые слова: многолетнемерзлые породы, газовые гидраты, поровый лед, прочность, теплопроводность, газовая проницаемость, электрическое сопротивление, метан

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