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Analysis of the Conditions of Mechanical Destruction of Clathrate Hydrates Using the Graphoanalytical Mohr Method

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Abstract

The article considers the mechanisms of mechanical destruction of clathrate hydrates. To determine the conditions of destruction, the hydrate is considered as a quasi-brittle body with insignificant plastic deformation during destruction. The possibility of using Mohr's graphoanalytical method to determine the critical values of tangential stresses arising under complex-stressed volumetric action during destruction by the method of pressure reduction (depression dissociation) is shown. The application of Freudenthal's theory with the presence of a critical defect in the material as a basis for forming an understanding of the relationship between the strength of the material and defects and the mechanics of hydrate destruction as a quasi-brittle material is also considered.

Keywords: Clathrate hydrates; Quasi-brittle material; Mohr-Coulomb theory; Mohr method; Freudenthal theory

1. INTRODUCTION

Recently, more and more works [1–10] devoted to the destruction of clathrate hydrates (hereinafter referred to as simply hydrates) have been published. Natural hydrates contained in the subsoil, when destroyed, are a huge source of natural gas [5]. At the same time, when transporting natural gases through pipelines, man-made hydrates can form in areas with subsequent changes in the cross-section completely closing the pipeline, and their destruction is necessary to ensure gas transportation [6]. The formation of natural and man-made hydrates is fundamentally similar, but has a number of differences, and their dissociation differs due to the limited possibility of their destruction due to technological and other conditions.

Technogenic hydrates formed in various sections of the pipeline are an undesirable phenomenon requiring measures to control, to prevent their formation and to destroy them when they appear. In principle, hydrates dissociate when the pressure decreases, the temperature increases, or chemical components are introduced into the system [5]. At the same time, the depression method of hydrate destruction is not always applicable from the point of view of technology and operational features of the pipeline systems themselves [6]. Often, the pressure reduction is designed not for mechanical destruction, but for dissociation (decomposition), although one can also find in the literature a description of mechanical destruction accompanying the process of decomposition of a gas hydrate by reducing pressure.

The purpose of this work is to analyze the possibility of realizing the process of mechanical destruction of clathrate hydrates by reducing the pressure and the possibility of determining the necessary and sufficient conditions of destruction based on the graph-analytical method of Mohr using the Mohr-Coulomb theory.

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2. BASIC PROVISIONS

Based on studies of mechanical and physical properties, clathrate hydrates were classified as brittle materials [9–11]. However, literature analysis [9–15] suggests that, based on their mechanical characteristics, they can be classified as quasi-brittle materials with their inherent brittle failure, preceded by conditional (minor) plastic deformation. It should be noted that the conditions for the formation of hydrates are determined by several factors, including the thermobaric conditions of systems containing a certain type of medium of varying saturation (concentration) of its components (particles of water, gas, condensate, etc.). Thus, at a certain temperature and pressure, a hydrate is formed from a certain initial concentration of a mixture of a hydrate former and water.

Depending on the pressure difference between the pressure P of hydrate formation and P_s of the surrounding medium, the hydrate may be mechanically destroyed or its integrity may be maintained. When the difference is less than or equal to the tensile strength of the hydrate σ,

$$\frac{P-P_s}{\sigma}=\frac{\Delta P}{\sigma}\le 1, \tag{1}$$

its mechanical integrity is preserved. If

$$\frac{\Delta P}{\sigma}>1 \tag{2}$$

the hydrate, according to Hooke's law, is mechanically destroyed.

The tensile strength σ is a function of the internal morphology of the hydrate, the main parameter of which is its density ρ_h:

$$\sigma=f_1\left(\rho_h\right). \tag{3}$$

In turn, the density depends on many other factors: the composition of the hydrate-forming agent, the degree of filling of the cavities of the crystal lattice, hydrate number, pressure and temperature of hydrate formation, etc. [2].

In addition, the internal morphology of hydrate φ is characterized by intercrystalline energy interactions that unite and bind hydrate crystals together. As a first approximation, these interactions can be expressed through the basic parameters σ and ρ_h:

$$\phi=\frac{\sigma}{\rho_h}. \tag{4}$$

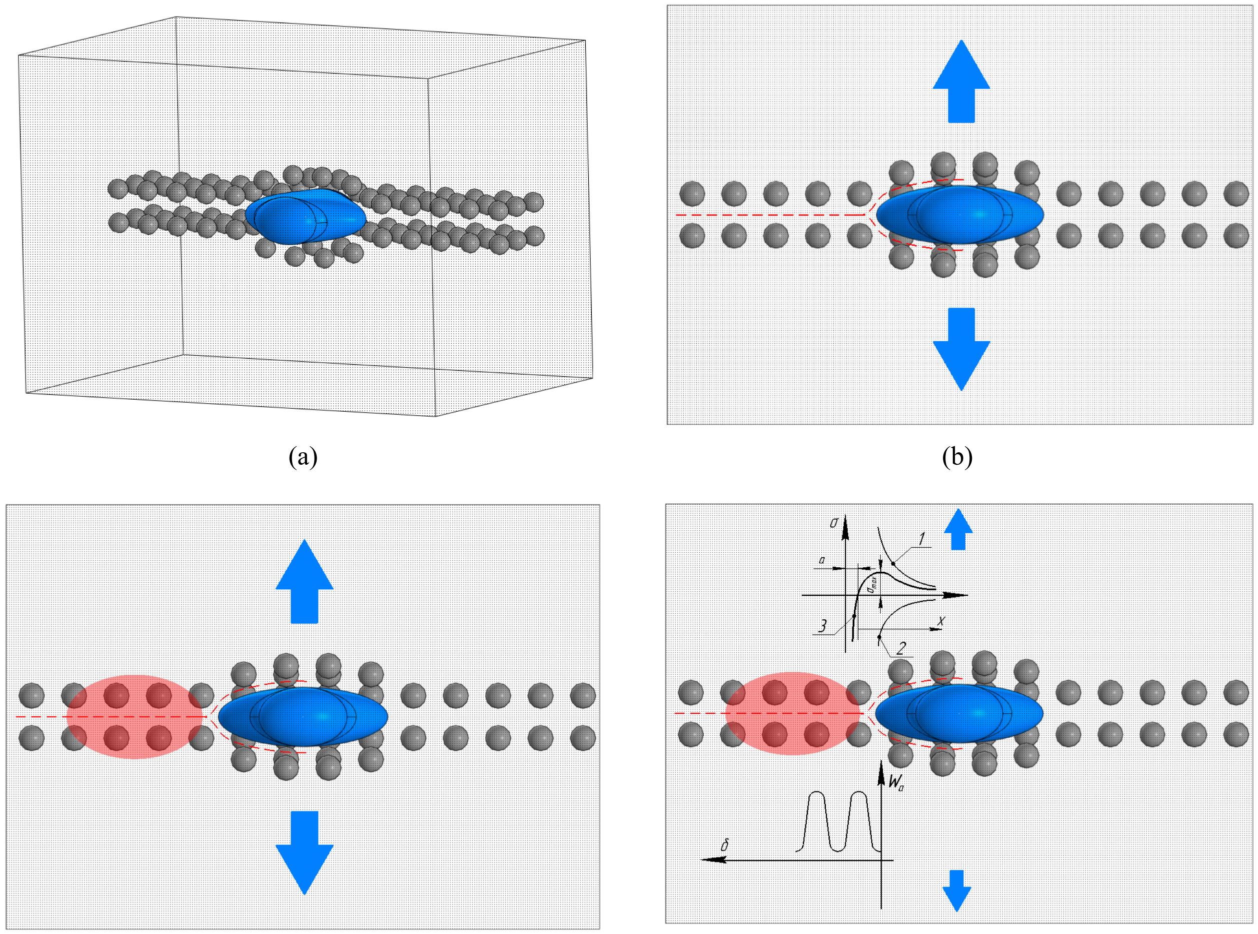
The value φ has the dimension of kJ/mol and reflects the specific energy of intercrystalline bonds of hydrate.

In terms of conditions, this is similar to the crystallization process of solids known today. In this case, the conditions for the formation of hydrates—pressure P and temperature T—are conditionally equilibrium for the existence of a solid phase, and the rate of formation from a gaseous (liquid) medium into a solid one assumes high values and the formation of a finely dispersed structure. It is also worth noting that polymorphism is characteristic of hydrates, as well as metals and alloys [5,16–20]. By reducing the temperature, it is possible to achieve a change in the structure of the solid phase of the hydrate to a more favorable structure of the equilibrium state for new conditions. It is worth noting that polymorphism manifests itself in hydrates even with a change in pressure (increase) [16–20], hence it can be concluded that with a further increase in pressure, the hydrate is rebuilt into a more energetically favorable structure.

All this makes it clear that in the volume-stressed state of the hydrate under compression for the purpose of mechanical destruction, the supplied energy will be spent on restructuring the hydrate, forming a structure stable in new thermobaric conditions. In this case, the finely dispersed structure will block the growth of cracks and the movement of defects, since its boundaries are barriers. It is also worth noting that the transition from the gaseous to the solid phase allows for the production of denser structures due to increased pressures. This is where the understanding of the use of the depression method of hydrate destruction comes from, since the process is accompanied by a decrease in pressure, which ensures the occurrence of tensile forces throughout the entire volume of the hydrate (volume-stressed state), thereby causing tensile normal stresses.

Hydrates, as was said above, can be classified as quasi-brittle materials, which also means that they work well under compressive loads and poorly under tension. Thus, the hydrate in equilibrium thermobaric conditions does not experience any effects, and the structure does not have any stress states, except for some residual ones appearing during the formation of crystals at a critical speed. For possible mechanical destruction, it is assumed that the pressure should be reduced to values ensuring the onset of hydrate decomposition, since the conditions of equilibrium existence change. In this case, continued pressure reduction can lead to the occurrence of a volume-stressed state of the hydrate with tensile stresses acting over the entire surface in all planes. In this scenario, if the pressure increases after the system has already adopted an energetically favorable structure, a further increase in pressure will also lead to the occurrence of a volume-stressed state, but with compressive forces. However, as is known [21–23], quasi-brittle materials work worse in tension than in compression, especially under conditions of a volume-stressed state.

When forming a hydrate, especially in a pipeline, in addition to pressure during the formation process, the flow action force and adhesion also affect, which cause a



(a) (b)

(c) (d)

Fig. 1. Structural defect in a hydrate similar to a microcrack: (a) hydrate unit cell with an internal volumetric structural defect; (b) interatomic propagation of a microcrack in one of the planes (opening of a structural defect); (c) pre-fracture zone of a hydrate (characteristic of quasi-brittle materials); (d) dependences of forces on the interatomic distance and the energy required for crack growth tper an interatomic distance, where *1* is the curve of the attractive force of atoms, *2* is the curve of the repulsive force of atoms, 3 is the curve of the resulting force, *a* is the interatomic distance of the compound.

complex stress state from a complex-loaded effect, which also add tangential stresses from the action of a shear effect from the flow pressure force and resistance to this effect from adhesion (shear) [24].

When a hydrate is destroyed by reducing pressure, two processes will occur simultaneously: crack formation along areas with maximum tangential stresses, their propagation, opening and, at the same time, decomposition of hydrates, with the two phenomena mutually accelerating each other. Being classified as quasi-brittle materials, hydrates will be sensitive to all types of loading where tensile stresses can occur, i.e., bending, rupture, etc.

3. RESULTS AND DISCUSSION

It is proposed to use Freudenthal's theory [25] for the destruction of hydrate as a quasi-brittle material with the presence of a critical defect in the material as its basis, associated with one of the hypotheses, the Weibull distribution [26], based on three other hypotheses:

- Frechet distribution of the largest defects [27];
- no interaction of adjacent defects and their sparse distribution;
- sharp microcracks (or defects) of the Griffith's type.

This theory shows the relationship between the strength of the material and defects and allows us to understand the mechanics of destruction of hydrate as a quasi-brittle material.

At the same time, in the presence of local defects that have critical values, acting as microcracks, crack propagation and its opening, suggests the presence of a plastic zone in front of the crack tip—the so-called local zone of plastic deformation (Fig. 1). Due to the structural features of the hydrate, from the point of view of crack propagation, its growth will proceed according to the principle of least resistance and will destroy either hydrogen bonds (6–160 kJ/mol) or Van der Waals (1–30 kJ/mol). When the hydrogen bond energy values are lower than the Van der Waals bond energy in the hydrate compound, the crack will grow along the hydrogen bonds between the hydrate molecules. If it is energetically more favorable for the crack to pass through the hydrate cluster molecule (when the hydrogen bond energy value is higher than the Van der Waals energy value), destroying the bond between the gas and water molecules. In this case, the hydrate as a quasi-brittle material has a pre-fracture zone characterized by a large length and width, with the length exceeding the width, thereby acquiring an elliptical (elongated) shape (Fig. 1c). It is worth noting that near the pre-destruction zone, there is practically no "hardening", and it can be considered that there is no plastic or strengthened nonlinear zone, which is characteristic mainly of elastic materials and partly of brittle ones.

The process of crack growth should be accompanied by the release of gas from the hydrate in the case of the crack front passing between the water molecules and gas molecules, destroying the bond between them formed by the Van der Waals forces. It should be taken into account that the environment also often contains gas and water molecules, which can shift the equilibrium conditions, thereby making it possible to form new hydrates along the newly formed hydrate surfaces behind the crack front, thereby connecting these surfaces (essentially, "collapsing" the crack).

And the crack itself increases the distance between the conjugated associates, breaking their bond. When an interatomic crack propagates, each compound is a barrier (a barrier of propagation energy or activation, or opening) in which more energy is required for the crack to develop, therefore the energy for the crack front propagation has the shape of a sinusoid. As can be seen from Figure 1d, to break the bond between atoms at the crack front, stresses σ must arise that exceed the theoretical strength of the adhesion of atoms $\sigma_{\max}$:

$$\sigma > \sigma_{\max} \tag{5}$$

The resulting stress must provide energy W sufficient to enable the growth of a crack by one atomic distance, exceeding the activation energy barrier W_a.

In this case, the formed hydrate may have zones free of defects (vacant zones) that can be filled with water molecules [28] reducing stresses arising near these zones, thereby stabilizing defects below their critical values, which ensures the stability of the hydrate. However, despite the stabilizing effect of such filling, stresses still remain and this element should be considered as a structural defect that reduces the strength of the hydrate.

It is worth noting that when forming an understanding of the destruction of hydrate as a quasi-brittle material, it is necessary to take into account that the existing approaches to the study and theory of the destruction of quasi-brittle materials in fracture mechanics are based on the behavior of materials during energy-releasing (exothermic) destruction, and the theory of crack propagation is also based on the mobility of atoms and the loss of bonds between atoms. In this case, the hydrate absorbs energy during decomposition, thereby creating conditions (a decrease in temperature at constant pressure) under which the existence of a solid phase and the possibility of forming a hydrate of the corresponding structure at the crack front and along its entire length at the corresponding interatomic distance are in equilibrium.

It is also worth noting that the conditions for the formation of hydrates in the presence of structural defects allow the formation of new bonds between water molecules at the sites of defects when exposed to pressure (especially under volumetric compression), thereby forming a denser crystalline structure and, thereby, increasing its strength.

Hydrate destruction occurs along areas with maximum tangential stresses, along which cracks nucleate, grow (propagate) and open. Of the existing theories of strength, the Mohr-Coulomb theory and the graphoanalytical method based on it with the construction of Mohr circles most accurately allow one to describe and determine the value of the arising tangential stresses for quasi-brittle materials. The only drawback of this theory is that this method does not take into account all three components of stress, since it is based on a plane construction taking into account the largest and smallest values of normal stresses arising as a result of volumetric action when decreasing (increasing) pressure. The result of such a calculation is an error of 10–15% of the real value, which is considered acceptable for engineering calculations. When constructing Mohr's circles, it is possible to determine the value of the arising tangential stresses on various sites. Including critical values from the action of destructive effects of the volumetric stress state of tensile and compressive forces.

It can be assumed that with a depression effect on the formed hydrate, it will be in a volumetric stress state, while the normal stresses on the main sites will have the following condition:

$$\sigma_1 > \sigma_2 \geq \sigma_3, \tag{6}$$

since the hydrate is constrained in two planes and by the critical pressure at the point of maximum narrowing of the flow cross-section as well as by the principal stress σ_1. In addition, the pressure of the flow tending to shift the hydrate in the direction of the gas flow is also considered. Thus, that part of the condition where it is reflected that σ_2 and σ_3 can be equal is an ideal condition for the uniform hydrate formation, which is difficult to achieve in laboratory conditions and has an extremely low probability in real operating conditions. However, it is possible that the difference between them will tend to zero (Fig. 2), which indicates that it is difficult to predict the orientation of the plane with maximum stress. At the same time, it is still possible to determine the maximum value of the limiting tangential, normal and shear stresses for a plane not parallel to any of the principal stress directions.

As mentioned earlier, the hydrate under depression action (due to the decrease in stress causing the appearance of tensile forces throughout the volume) is not only in a volume-stressed state, but also in a complex loading state, since it experiences shear forces along the adhesive interface in addition to the volumetric stress. Thus, of the existing methods for analyzing the limit state of the hydrate under the effect of pressure changes, the Mohr-Coulomb theory is suitable (Fig. 3).

Using the limit curve, it is also possible to construct a family of Mohr's limit circles for hydrates with the determination of the necessary values of σ_1 and σ_3 corresponding to the limit state for different stress states and, as a consequence, the conditions for the occurrence of maximum permissible shear stresses for the destruction of the hydrate. The hydrate crystal lattice is ice-like and many properties of hydrates are similar to hexagonal ice [5,28]. In addition, at low temperatures with an excess of the water component over the absorbed molecules (especially in natural conditions), the joint existence of hydrate-ice deposits (clusters) is possible. Below in Figure 4 are presented Mohr's circles and the limit envelope for ice at a temperature of –5 °C, constructed based on an analysis of the literature [29–32] with experimental data. In this case, under uniaxial compression and tension, destruction occurs along areas with maximum shear stresses located at angles of ~45° from the direction of force application or principal stress. Thus, the limiting values of shear stresses under uniaxial compression and tension obtained using graphical analysis of Mohr's circles from Figure 4 are equal, respectively, to $\tau_{UC} \sim 2.1 \pm 0.5$ MPa and $\tau_{UT} \sim 0.52 \pm 0.06$ MPa (τ_{UC} is ultimate tangential stress under uniaxial compression, τ_{UT} is ultimate tangential stress under uniaxial tension determined with the help of Mohr's circle).

Figure 5 shows Mohr's circles and the limiting envelope for methane hydrate at a temperature of –5 °C, constructed based on an analysis of experimental data [11,12]. In this case, under uniaxial compression and tension, failure also occurs along areas with maximum shear stresses located at angles of ~45° from the direction of application of force or principal stress. In this case, the limiting values of shear stresses under uniaxial compression and tension obtained using a graphical analysis of Mohr's circles from Figure 5 are, respectively, equal to $\tau_{UC} \sim 3.65 \pm 0.45$ MPa and $\tau_{UT} \sim 1.23 \pm 0.1$ MPa.

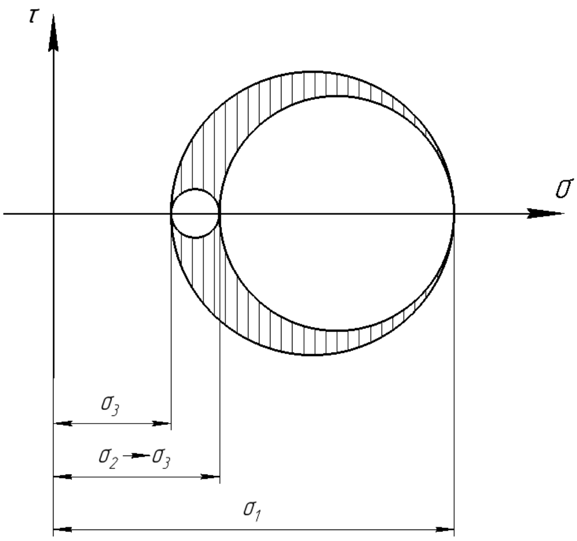


Fig. 2. Mohr's circles of the three principal stress directions of the family of hydrate sites.

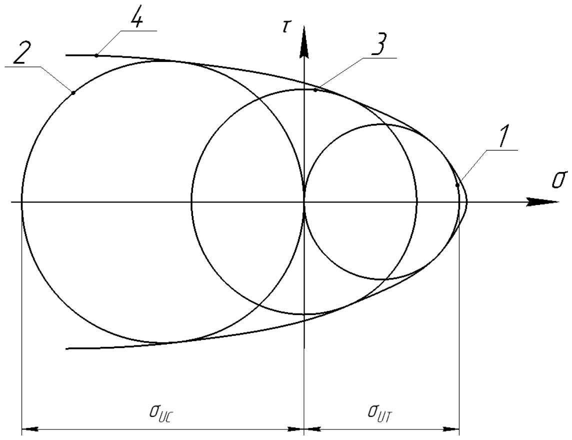


Fig. 3. Mohr's circles and the limit envelope for hydrate, where: *1* is the Mohr's limit circle for tension; *2* is the Mohr's limit circle for compression; *3* is the Mohr's limit circle for shear; *4* is the limit envelope. σ_{UC} is the ultimate normal stress in uniaxial compression, σ_{UT} is the ultimate normal stress in uniaxial tension.

Using the obtained limit envelopes for ice (Fig. 4) and for methane hydrate (Fig. 5), we can determine the conditions under which they would not have sufficient strength, leading to their destruction under external forces. If the constructed Mohr's circle under these forces based on the values of the principal stresses σ_1 and σ_3, intersects its limit envelopes, respectively, then the destruction of ice or hydrate will occur, but if the Mohr's circle is located inside the limit envelopes, then the ice or hydrate will have sufficient strength, and they will not be destroyed. It is worth noting that hydrates are sensitive to all types of dynamic and cyclic loading under given conditions of the system dynamics. In this scenario, the amount of work required to open the crack will depend on the speed and external conditions of the hydrate's existence. The crack can be closed

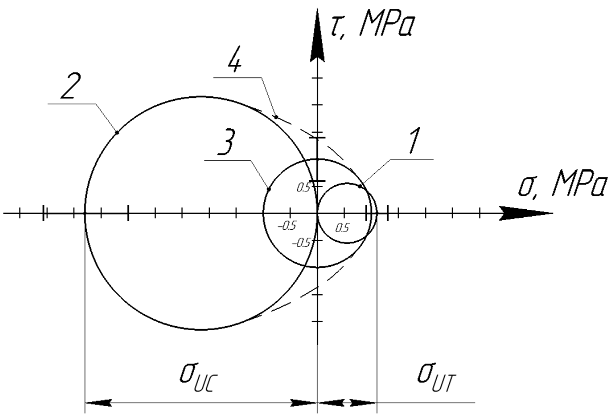


Fig. 4. Mohr's circles and the limit envelope for ice, where: *1* is the Mohr's limit circle for tension; *2* is the Mohr's limit circle for compression; *3* is the Mohr's limit circle for shear; *4* is the limit envelope. σ_{UC} is the ultimate normal stress in uniaxial compression, σ_{UT} is the ultimate normal stress in uniaxial tension for ice [29–32].

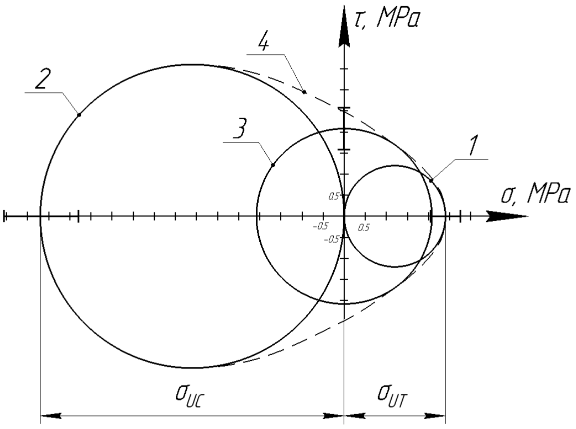


Fig. 5. Mohr's circles and the limit envelope for methane hydrate, where: *1* is the Mohr's limit circle for tension; *2* is the Mohr's limit circle for compression; *3* is the Mohr's limit circle for shear; *4* is the limit envelope. σ_{UC} is ultimate normal stresses in uniaxial compression, σ_{UT} is ultimate normal stresses in uniaxial tension for methane hydrate [11,12].

by newly formed crystals along its formation path, as the hydrate's destruction involves heat absorption, which also creates conditions for reducing the temperature and achieving equilibrium for the solid phase at the appropriate pressure.

To form an understanding of the mechanisms of mechanical destruction of various clathrate hydrates, it is necessary to study their physical and mechanical properties in more detail and construct a family of Mohr's circles for various limiting states. Also, to understand the discrepancy between real values and those obtained using the Mohr-Coulomb theory based on the graphoanalytical analysis, it is necessary to consider the destruction process from the standpoint of fracture mechanics for quasi-brittle materials based on the Freudenthal's theory based on the Weibull distribution hypotheses. This will make it possible to create a physical and mathematical model that accurately describes the conditions of hydrate destruction, avoiding the errors inherent in values obtained through calculations based on the Mohr-Coulomb theory. The emergence of an understanding of the process of mechanical destruction of hydrates and the formation of a theory will allow a better understanding of the mechanisms occurring during depression destruction and the formation of more effective approaches and technological support for their mechanical destruction.

4. CONCLUSION

In this work, Mohr's circles and the limit envelopes for ice and methane hydrate are constructed for the first time based on the analysis of experimental data available in literature. Hydrates are also considered for the first time as quasi-brittle materials and the theory of crack propagation in a quasi-brittle material with subsequent destruction of the hydrate formation is used. The conditions for crack opening and the direction of crack front propagation are considered. Using Mohr's circles and the Mohr-Coulomb theory, the values of shear stresses for ice and methane hydrate at a temperature of –5 °C are determined for areas with maximum values of shear stresses oriented at an angle of ~45° relative to the direction of force application or the principal stress under uniaxial compression and tension. Based on the proposed method, it is possible to construct a series of Mohr's circles for different limiting states of crystalline structures of ice and hydrates, upon reaching which their mechanical destruction occurs. On the basis of these results it is possible to determine the necessary baric influence on hydrates in order to destroy them.

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Анализ условий механического разрушения клатратных гидратов с использованием графоаналитического метода Мора

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Аннотация. В статье рассмотрены механизмы механического разрушения клатратных гидратов. Для определения условий разрушения гидрат рассматривается как квазихрупкое тело, имеющее незначительную пластическую деформацию при разрушении. Показана возможность применения графоаналитического метода Мора для определения критических значений касательных напряжений, возникающих при сложнонапряженном объемном воздействии при разрушении методом снижения давления (депрессионной диссоциации). Также рассмотрено применение теории Фрейденталя с наличием критического дефекта материала в качестве основы для формирования понимания связи между прочностью материала и дефектами и механики разрушения гидрата как квазихрупкого материала.

Ключевые слова: клатратные гидраты; квазихрупкий материал; теория Мора-Кулона; метод Мора; теория Фрейденталя