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DEVELOPMENT OF A MATHEMATICAL MODEL FOR GAS HYDRATE SYNTHESIS AND DISSOCIATION PROCESSES IN A GAS PIPELINE UNDER MICROWAVE RADIATION

The object of research is the heat and mass transfer processes that occur on the surface of a gas hydrate granule moving in a gas pipeline.

The research solved the problem of the oil and gas industry – the fight against the formation of gas hydrates (GH) in hydrocarbon production and transportation systems. A promising approach to the intensification of hydrate dissociation by using microwave electromagnetic radiation is proposed. The features of the results obtained consist in the development of a mathematical model that combines two processes of synthesis and dissociation of gas hydrates, taking into account hydrodynamic processes in the pipeline. The main differences of the proposed mathematical model are: simultaneous consideration of two mutually opposing processes of synthesis and dissociation of gas hydrate on the surface of the granule; these processes occur during the movement of the gas hydrate granule inside the gas pipeline; heating of the gas hydrate granule occurs as a result of its absorption of direct and reflected multimode microwave electromagnetic radiation.

A complex approximation dependence was obtained for determining the required emitter power $P_w = 124C_{gh}/R^{1.7}$*, which allows one to quickly calculate the required minimum magnetron power to combat hydrate formation.*

It was established that for the destruction of small particles (<0.01 mm) even if their concentration is low (0.01–0.02%), a relatively high-power emitter (3–5 kW) is required. The same was observed in the case of GH accumulation to high concentrations (>0.1%) with particle sizes of 0.05–0.1 mm. Therefore, the most advantageous strategy for combating GH accumulation is its active destruction when it reaches sizes of 0.02–0.05 mm, which do not harm the course of the main technological process.

The obtained dependences are the basis for improving gas preparation technologies under hydrate formation conditions by using microwave radiation energy.

Keywords: *gas hydrates, ultrahigh-frequency electromagnetic radiation, dissociation, mathematical modeling, gas pipeline, energy efficiency, heat balance.*

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1. Introduction

The problem of hydrate formation in the processes of extraction and preparation of hydrocarbon raw materials in various sections of pipelines is faced by oil and gas companies in different regions of the world [1]. Most often, blockages of gas pipelines occur in the winter period due to significant cooling of the gas flow moving in the pipeline. Hydrate formation can occur in all gas pipelines, except those that transport gas with a water vapor dew point below the minimum operating temperature [2]. Carrying out work to eliminate these complications dramatically increases the cost of operating wells and hydrocarbon collection systems [3]. Traditional methods of combating hydrate formation, such as the introduction of chemical inhibitors, pressure reduction or external heating, often turn out to be energy-intensive, environmentally hazardous or not fast enough [4]. In this context, the use of microwave (MW) radiation is considered one of the most promising technologies due to the possibility of non-contact volumetric heating of the medium, high energy transfer rate and the ability to selectively affect components with high dielectric losses [5]. One of the effective tools for studying such phenomena is mathematical modeling of heat and mass transfer processes. However, the variety of physical models of the formation and dissociation of gas hydrates (GH) leads to different approaches to the mathematical description of the processes [6]. Creating an adequate mathematical model capable of describing this complex of multiphysical processes – from the propagation of electromagnetic waves in a heterogeneous medium to phase transitions and non-stationary heat and mass transfer processes – is a difficult scientific task that faces serious limitations of modern numerical methods, and the use of neural networks requires their training on high-precision mathematical models or experimental data [7].

The propagation of electromagnetic energy in such a system occurs in the form of certain types of waves (modes), each of which has its own field structure and attenuation coefficient [8]. Although the modes themselves are characteristic of a coaxial line, which greatly simplifies the mathematical model (the frequencies can be adjusted within wide limits, there are almost no losses on the surface of the pipelines, but only in the GH). In fact, TEM waves (characteristic of a coaxial line) are not used in our case. When modeling microwave dissociation in a pipeline, it is necessary to take into account that the metal wall of the pipe acts as a cylindrical waveguide [9]. The propagation of electromagnetic energy in such a system occurs in the form of certain types of waves (modes), each of which has its own field structure and attenuation coefficient [8]. Filling the pipeline with dielectric clathrates (for example, gas hydrate) changes the absorption intensity of various modes in the waveguide. The mathematical model must take into account the dynamic change in the concentration of gas hydrate during its formation or dissociation in the pipeline, which affects the local radiation power [10]. If there is a valve, constriction or diaphragm at the ends of the pipeline, which sharply reduces its cross-section, this can lead to reflection of electromagnetic waves [11]. The mathematical description of the process of dissociation of gas hydrates under the influence of microwave radiation is based on a combination of Maxwell's electrodynamic equations and the system of heat and mass transfer equations in a multiphase medium [12]. Unlike traditional surface heating, where the heat flow is directed from the phase interface, microwave influence generates internal energy sources, the intensity of which depends on the local electric field strength and the phase composition of the medium [13].

In [14], the heating of a cylindrical volume (in the well pipeline) completely filled with gas hydrate is considered. The supply of microwave EM radiation is carried out using a coaxial cable, due to which volumetric heat sources are formed in the gas hydrate. The mathematical model proposed by the authors does not take into account the propagation of EM waves along the pipeline axis, and also does not contain the separation of EM waves into modes. The most critical aspect of the implementation of mathematical models of microwave dissociation is the choice of a numerical method of integration over time [15]. Indeed, due to the large difference in speed scales (the dynamics of the EM wave vector at the speed of light and the movement of GH granules in the pipeline at speeds up to 10 m/s); the consideration of high-frequency waves in macroobjects can lead to large expenditures of computer time. The author recommends using geometric optics methods, which in this case can give a good approximation. In this way, it is possible to avoid considering microobjects and save time (machine time) on calculating macroobjects. This approach is implemented in our mathematical model.

Explicit Runge-Kutta methods (in particular, 4th order) are promising for modeling gas hydrate dissociation processes. They are a standard for many engineering problems due to their simplicity of implementation and high accuracy for non-rigid systems [15].

In paper [16], a comparison of the Euler and Runge-Kutta 4th order methods was made. The Runge-Kutta method was recognized as a much more effective and powerful tool. The Euler method, although simple to implement, turned out to be the least accurate (0th order of accuracy) and requires extremely small steps to achieve acceptable results.

In paper [17], a one-dimensional mathematical model for simulating the physical processes of methane hydrate decomposition in porous media is presented. The mathematical apparatus includes the energy, gas and liquid conservation equations, as well as the thermodynamic equilibrium equation between pressure (P) and temperature (T). The authors used an implicit finite-difference scheme to solve the system of equations, which allows for an accurate description of the appearance of new phases and boundary conditions. The applied method has an accuracy of 1–2 (versus 4 in the Runge-Kutta method), but is absolutely stable, which is important for sharp changes in thermophysical parameters (for example, temperature or pressure).

Studies of literary sources indicate the need to improve the mathematical model of non-stationary thermal regimes of one-dimensional elements taking into account the hydrodynamics of flow in the pipeline cross-section in order to expand the scope of its application.

The object of research is the heat and mass transfer processes that occur on the surface of a gas hydrate granule moving in a gas pipeline.

The aim of research is to develop a mathematical model of the processes of synthesis and dissociation of gas hydrates in a gas pipeline under the action of microwave radiation. This will make it possible to calculate the required minimum magnetron power to combat hydrate formation at a specific facility.

Research objectives:

1) to develop a mathematical model for calculating the transient processes of synthesis and dissociation of gas hydrate granules under the action of microwave radiation, taking into account the hydrodynamics of the gas flow, which will allow predicting changes in their size and concentration along the gas pipeline;

2) to establish quantitative relationships between the power of EM radiation and the parameters of hydrate particles to select the operating modes of the emitter and ensure the hydrate-free operation of pipelines.

2. Materials and Methods

In the mathematical modeling of heat and mass transfer processes on the gas pipeline section, the finite element method was used, in particular, the entire length of the pipeline was divided into 100 sections and the entire calculation was repeated for each individual section. Since transient processes also occur in time coordinates, the Runge-Kutta method of the 4th order was used for the time integration of differential equations.

It was assumed that gas hydrate is evenly distributed over the cross-section of the gas pipeline in the form of microgranules, the size of which, and therefore the specific mass of the hydrate, will depend on the processes that prevail on this section of the gas pipeline.

The analysis of the results of modeling heat and mass transfer on the surface of a gas hydrate granule in the gas pipeline is complicated by the fact that the process is heterogeneous, accompanied by phase transitions, the release of latent heat and a change in the size of the granule itself during its transportation in the gas pipeline. To interpret the data sets obtained during mathematical modeling, the following specialized analysis methods were used:

1. *Spatial and boundary analysis methods.* These methods allow to assess the local intensity of heat and mass exchange processes directly at the gas-hydrate interface and to understand which macroflow factors in the gas pipeline most affect the state of the granule. In particular, determining the Nusselt criterion (Nu) allows to calculate the heat transfer coefficient and mass transfer intensity on the granule surface and identify zones of intensive hydrate growth or decomposition.
2. *Integral methods of balance and thermodynamic analysis.* They assess the behavior of granules in the gas flow as a single system. The method of integral mass and energy balances: Comparison of the total amount of heat removed by the gas flow with the amount of heat released as a result of the exothermic phase transition reaction and entering the granule as a result of the absorption of microwave radiation allows to determine the direction of the gas hydrate synthesis or dissociation process and observe the law of conservation of energy.
3. *Methods of analysis of non-stationary fields and kinetics of geometric changes.* Since the gas hydrate granule grows or collapses during movement in the gas pipeline, its size is constantly changing, which affects the hydrodynamics of the flow around it. The method of tracing the interphase boundary allows to perform a quantitative analysis of the evolution of the granule size over time and to assess how the change in the surface area of the phase contact affects the total heat flow.

To perform mathematical modeling tasks, the Pipe3 computer program, written in the QBASIC language, was developed. To perform calculations, the entire length of the pipeline is divided into 100 sections. The calculated time step is 1/104 of the gas residence time in the gas pipeline. Differential equation (6) is solved by the 4th order Runge-Kutta method.

Structurally, the computer program provides for:

1. Declaration of modules, procedures, arrays, variables.
2. Specification of input data, constants and initial state.
3. Execution of the calculation cycle of the mathematical model equations over time with a nested cycle along the length of the gas pipeline.
4. Output of the calculation results to the screen and to the file pipe1.dat. This file is read by the Excel program, which allows them to be processed and analyzed.

3. Results and Discussion

3.1. Mathematical model of the synthesis and dissociation of gas hydrate in a gas pipeline

3.1.1. Synthesis of gas hydrates (GH)

Even in the presence of suitable thermobaric conditions, a seed crystal, water droplets or dust particles are required to initiate hydrate formation. After intensive throttling on the regulating body, at least there will be drops of water in the gas pipeline. Therefore, it was assumed that such a microcrystal (microgranule) of gas hydrate already exists (the worst case scenario) and it moves in the flow of cooled gas in the gas pipeline. It is obvious that the growth of this microgranule of the crystal occurred as a result of the removal of the heat of hydrate formation from its surface.

A physical model of the process of hydrate formation on the surface of a GH granule was considered, assuming the thickness of the GH crust (δ_{gh}) to be three to four orders of magnitude smaller than the size of the granule itself, Fig. 1.

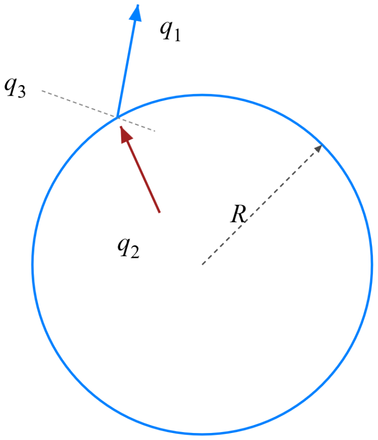

Fig. 1. Scheme of heat flows near the interfacial surface: q_1 – heat flow into the gas medium; q_2 – heat flow from microwave EM radiation; q_3 – heat release from the hydrate formation process; R – radius of the GH granule

For the GH formation, it is necessary to remove the heat of hydrate formation (q_3) into the gas medium (q_1). Since the GH is under the influence of microwave electromagnetic radiation (EM), internal volumetric heat sources (q_2) operate in its volume, which heat the granule, worsening the conditions for hydrate formation. The intensity of the formation/dissociation of the GH crust on the granule surface is determined by the heat balance (q_3), Fig. 1.

To calculate the hydrate formation on the granule surface, a system of equations was used, which consists of the equation of the granule heat balance, W

$$q_3 = q_1 - q_2, \tag{1}$$

where q_3 – heat input from the hydrate formation process, W; q_1 – heat removal to the cold gas medium, W; q_2 – heat supply of microwave radiation, W

$$q_3 = 4\pi R^2 m_{gh} \cdot r_{gh}, \tag{2}$$

where r_{gh} – heat of GH dissociation, J/kg; m_{gh} – specific mass quantity of gas hydrate formed, kg/(s · m^2); R – granule radius, m. Heat of cooling of the gas hydrate granule

$$q_1 = \alpha_1 4\pi R^2 \left(T_0 - T_g\right), \tag{3}$$

where α_1 – heat transfer coefficient near the interphase surface from the gas phase side, W/(m^2 · K); T_0 – gas temperature at the hydrate formation line, K; T_g – gas temperature in the gas pipeline, K

$$q_2 = q_v 4/3\pi R^3, \tag{4}$$

where q_v – specific volumetric heat source, as it arises as a result of the action of microwave radiation, W/m^3.

By substituting equations (2), (3) and (4) into 1, the rate of hydrate formation (if +) or dissociation (if –) is determined

$$m_{gh} = \frac{1}{r_{gh}}\left[\alpha_1\left(T_0 - T_g\right) - q_v\frac{R}{3}\right]. \tag{5}$$

Analysis of the obtained equation (5) shows that under the conditions of microwave radiation, with an increase in the radius of the granule, a moment will come when the mass of the formed hydrate will become equal to $m_{gh} = 0$ and the granule will stop growing. To clarify the time of occurrence of this moment, the initial and boundary conditions were determined.

The value of the radius of the gas hydrate granule is determined by its volume. The initial volume of the granule is given (V_0) – considering it as the embryonic center of hydrate formation. The rate of gas hydrate formation is determined from the equation

$$\frac{dV_{gh}}{d\tau} = \frac{m_{gh}}{\rho_{gh}}, \tag{6}$$

where τ – time, s; ρ_{gh} – the density of gas hydrate, kg/m^3. From the volume of the granule, it is easy to determine its radius, m

$$R = \sqrt[3]{\frac{3V_{gh}}{4\pi}}. \tag{7}$$

To calculate the heat transfer coefficients on the surface of water droplets sprayed in the gas, the similarity equation of the form [12, 13] was used

$$Nu = 2.0 + 0.6Re^{0.5}Pr^{1/3}. \tag{8}$$

This equation can be used within the values of the Reynolds criterion $0 < Re < 1000$. The Reynolds criterion in equation (8) is determined by the mutual velocity of the liquid drop and the gas flow in which it is located. Revealing the values of the criteria, the heat transfer coefficient, W/(m^2 · °C)

$$\alpha_1 = \frac{\lambda_g}{R}\left(1.0 + 0.424\left(\frac{VR}{\nu_g}\right)^{0.5} Pr^{1/3}\right). \tag{9}$$

The density of the gas moving inside the gas pipeline over a section 25 m long is taken as a constant value. It is determined by the composition of the gas, its temperature and pressure.

3.1.2. Dissociation of gas hydrates

The intensity of the EM field in which the GH granule is located at a certain point in time depends on the distance to the EM radiation source, which is located in the middle of the working section of the gas pipeline. Along the gas pipeline, the gas hydrate granules move at the gas velocity (V_g), therefore, at the initial time point ($\tau = 0$) the particle coordinate $x = -l/2$. After a time interval $\tau = l/(2V_g)$ the particle will reach the position $x = 0$. The particle will leave the pipeline after reaching the coordinate $x = l/2$ and completing the time interval $\tau = l/V_g$.

Summarizing the above, the granule coordinate is expressed by the formula

$$X = \tau V_g - \frac{l}{2}. \quad (10)$$

To control the amount of gas hydrate in the gas pipeline, an indicator of the GH concentration in the pipeline – C_{gh} was introduced. If its value $C_{gh} = 0$ – there is no hydrate in the gas pipeline. If $C_{gh} = 1$ – the gas hydrate has completely filled the entire internal space of the gas pipeline. Its quantitative value is defined as the ratio of the total volume of granules to the internal volume of the gas pipeline in which they are located

$$C_{gh} = \frac{N\cdot 4\pi R^3}{3V_{TP}}. \quad (11)$$

Given the constant number of granules ($N = \text{const}$) at any section of the gas pipeline, it is determined by the initial concentration ($C_{gh(0)}$) at the entry point ($x = -l/2$)

$$N = \frac{3V_{TP}C_{gh(0)}}{4\pi R^3}. \quad (12)$$

The volume of the pipeline in digital calculations is determined by its internal radius (R_B) and the calculated change in the length of the section (ΔX)

$$V_{TP} = \pi R_B^2\cdot \Delta X. \quad (13)$$

In a waveguide of circular cross-section (which is essentially a gas pipeline), only two types of electromagnetic waves can propagate: electric E_{mn}, in which $E_z \neq 0$, and $H_z = 0$, and magnetic H_{mn}, in which the components $E_z = 0$, $H_z \neq 0$. In a pipeline with a diameter equal to the critical one, only one type of waves propagates. As the diameter of the pipeline increases, the number of different types of electromagnetic waves (modes) that can propagate in it increases, in particular modes of the type H_{11}, E_{01}, H_{01} and others.

The condition for the propagation of electromagnetic waves of the H_{mn} and E_{mn} types in a pipeline of circular cross-section is the expression

$$\lambda < \lambda_{KP}^{mn}, \quad (14)$$

where $\lambda = \lambda_0/\sqrt{\varepsilon\mu}$ – the length of a plane uniform wave in an unlimited space filled with the same dielectric as the waveguide; ε, μ – are the relative dielectric and magnetic permeability of the dielectric that fills the pipeline; λ_0 – the wavelength in a vacuum; λ_{KP}^{mn} – the critical wavelengths for the H_{mn} and E_{mn} types.

High-frequency electromagnetic waves that propagate along the pipeline lose their power due to losses in the metal wall of the pipe and in the dielectric (hydrate) that fills the internal space. In the cylindrical coordinate system, the distribution density of volumetric heat sources is determined by the expression [14]

$$q = -\frac{1}{2}Re\frac{\partial}{\partial X}\left(\dot{E}_r\cdot\dot{H}_\varphi\right), \quad (15)$$

where *Re* – the symbol of the real part of the complex number; X – the spatial coordinate along the pipeline axis, m; $\dot{E}_r$ – the radial component of the electric field strength, V/m; $\dot{H}_\varphi$ – the azimuthal component of the magnetic field, A/m.

As can be seen from expression (15), the density of heat sources created in the medium interacting with the electromagnetic field is also a function of the electric field strength, which depends on the geometry of the system in which these waves propagate.

Considering a steel gas pipeline as a circular waveguide, the components of the electromagnetic field strength are determined from the solutions of Maxwell's equations by the formulas:

$$\dot{E}_{0r} = \frac{\dot{D}}{r}\ell^{-j\dot{\Gamma}X}, \quad (16)$$

$$\dot{H}_{0\varphi} = \frac{\dot{E}_{0r}}{\dot{Z}_C} = \frac{\dot{D}}{\dot{Z}_C r}\ell^{-j\dot{\Gamma}X}, \quad (17)$$

where D – the complex amplitude of the electromagnetic wave; Z_C – the wave resistance of the dielectric that fills the space of the pipeline, Ohm; j – an imaginary unit; $\dot{\Gamma}$ – the propagation constant of electromagnetic waves, m^{-1}; r, φ – spatial coordinates of the distance from the center of the pipeline and the angle, m and rad.

An expression was obtained to determine the power of local sources

$$q = \frac{2\alpha P_0}{\pi R^2}\ell^{-2\alpha X}, \quad (18)$$

where P_0 – the input power, W.

Taking into account the concentration of gas hydrate in the pipeline (C_{gh}), it was determined that the total attenuation coefficient of electromagnetic waves consists of two parts

$$\alpha = C_{gh}\alpha_{gh} + \alpha_{TP}, \quad (19)$$

where α_{gh} – for gas hydrate; α_{TP} – for a steel pipeline.

The attenuation coefficients due to losses in the metal walls of a circular waveguide were calculated by the formulas, m^{-1}:

- for waves of the H_{mn} type

$$\alpha_{TP} = \frac{Rs}{Z_C\cdot R\sqrt{1-\left(\frac{\lambda_0}{\lambda_{KP}^{mn}}\right)^2}}\left[\left(\frac{\lambda_0}{\lambda_{KP}^{mn}}\right)^2 + \frac{m^2}{\eta_{mn}^2 - m^2}\right], \quad (20)$$

- for waves of the E_{mn} type

$$\alpha_{TP} = \frac{Rs}{Z_C\cdot R\sqrt{1-\left(\frac{\lambda_0}{\lambda_{KP}^{mn}}\right)^2}}, \quad (21)$$

where R_S – the active surface resistance of the metal of the pipeline.

For hydrate, the attenuation coefficient was calculated by the formula

$$\alpha_{gh} = \frac{\pi}{\lambda_0}tg\delta\frac{Z_B}{Z_C}, \quad (22)$$

where Z_B – the characteristic impedance of the waveguide, Ohm.

Given the different diameters of gas pipelines, a common case may be the simultaneous existence of several EM radiation modes in the pipeline cross-section. Each mode propagates independently of the others – the principle of superposition. The most characteristic modes of EM waves that can propagate in a pipeline of circular cross-section are H_{11}, E_{01} and H_{01}.

Under the conditions of the research, the main factor of the thermal effect on gas hydrate is the exponential nature of the attenuation of each type of wave

$$P^{mn}_{(X)} = P^{mn}_{0}\ell^{-2\alpha^{mn}X}. \qquad (23)$$

To calculate the multimode regime, the initial power of each mode was determined

$$P^{mn}_{0} = P_{0}\frac{P^{mn}}{\sum\limits_{i=1}^{\psi} P^{mn}} = P_{0}k_{i}, \qquad (24)$$

where ψ – the total number of EM wave modes in this pipeline; P^{mn} – the EM wave power of a separate mode at a given constant value of $E = 1$, kV/cm; k_i – the power coefficient of the i-th mode.

Thus, knowing the power distribution between the modes, it is possible to determine the total power of EM waves at a distance X from the emitter at any distance

$$P_{(X)} = \sum\nolimits_{i=1}^{\psi} P^{mn}_{0}\ell^{-2\alpha^{mn}X} = P_{0}\sum\nolimits_{i=1}^{\psi} k_{i}\ell^{-2\alpha^{mn}X}. \qquad (25)$$

Reflection of EM waves from the ends of the pipeline: If the GH concentration is low, the EM wave does not have time to completely decay to the end of the pipeline. Then it is reflected and returns in the opposite direction. It was assumed that the process of reflection of the EM wave from the end of the pipeline occurs without energy loss.

Under the conditions of applying digital calculation methods, the reflected wave was calculated: from the left end of the gas pipeline

$$P_{(X)} = P_{0}\sum\nolimits_{i=1}^{\psi} k_{i}\ell^{-2\alpha^{mn}(L+X)}. \qquad (26)$$

And from the right end of the gas pipeline

$$P_{(X)} = P_{0}\sum\nolimits_{i=1}^{\psi} k_{i}\ell^{-2\alpha^{mn}(L-X)}. \qquad (27)$$

The resulting EM field power at each point of the internal space of the gas pipeline is equal to the sum of the powers of the direct and reflected (left and right) waves.

To avoid the accumulation of methane hydrate in the separator, the diaphragm in front of it is not used. In this case, the EM radiation is reflected only from one side – from the side of the throttling armature. This circumstance was taken into account when writing the computer program.

3.2. Results of calculating the dependence between the power of EM radiation and the parameters of hydrate particles

Input data for calculating the formation/dissociation of GH on the section of the gas pipeline from the throttling valve to the separator: hydrate-forming gas is methane; its thermal conductivity is 0.031 W/(m · °C); Prandtl criterion Pr = 0.734; gas temperature –30.1°C; hydrate formation onset temperature (at a given methanol concentration) –23.4°C; ambient temperature +16°C; gas pressure 37 bar; gas density 38.08 kg/m^3; gas velocity 4.287 m/s; internal diameter of the gas pipeline 257 mm; section length 25 m; gas hydrate density 913 kg/m^3; heat of GH dissociation r = 464 kJ/kg; calculated EM radiation modes H_{11}, E_{01} and H_{01}; EM wavelength 12.491 cm, characteristic values for hydrate are ε' = 3.75 and tgδ = 0.02.

The results of the calculation of the "growth" of the radius of GH granules along the gas pipeline section in the absence of EM radiation are shown in Fig. 2. The calculations show that in the absence of destructive radiation, hydrate particles of any size grow rapidly. Moreover, small particles grow more actively, since they have a larger total heat exchange surface area. In particular, during movement in the gas pipeline, particles with a size of 0.01 mm increased by 5 times, and particles with an initial size of 0.1 mm – by 2.5 times. In a real gas pipeline, particles will coalesce (coalescence), which will lead to the formation of particles of different sizes.

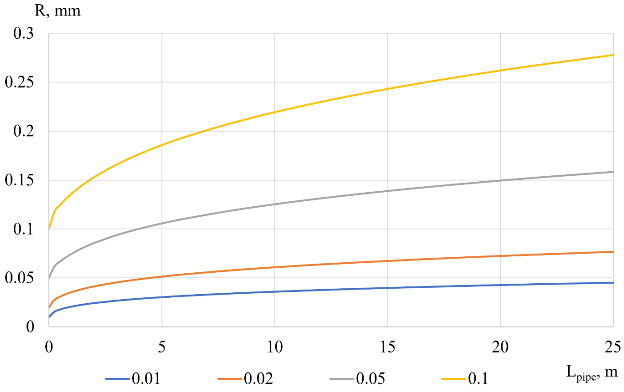

Fig. 2. Change in the size of gas hydrate particles along the length of the gas pipeline section in the absence of EM irradiation (the legend indicates the initial size of GH granules, mm)

In general, the increase in GH particles leads to an "avalanche" process of GH increase in the gas pipeline. Along with the growth of particles, the GH concentration in the pipeline also increases – Fig. 3. Thus, for particles with an initial size of 0.01 mm, the gas hydrate concentration at the end of the pipeline increases by 68 times, and for particles with a size of 0.1 mm – by 17 times. Thus, the increase in GH concentration is particularly strongly influenced by small particles, since at the same initial concentration their heat and mass exchange surface area is much larger than for large particles.

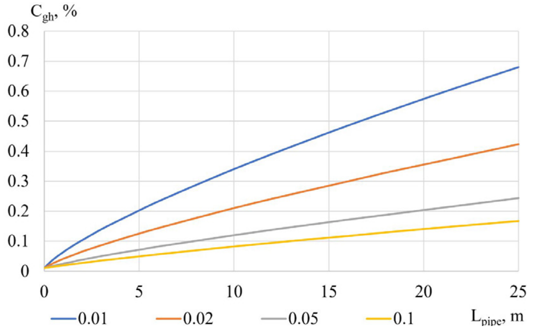

Fig. 3. Change in gas hydrate concentration along the length of the gas pipeline section in the absence of EM irradiation (the legend indicates the initial size of GH granules, mm)

Introducing EM radiation into the gas pipeline first leads to a slowdown in hydrate formation (1000–1500 W), then to an unstable state of concentration stabilization (1500–1800 W), and with a further increase in power (more than 1800 W) – to the destruction of existing GH, Fig. 4. The calculation results show that a further increase in the emitter power (2–3 kW) allows for faster decomposition of GH granules, but it no longer affects the final GH concentration (at the outlet of the gas pipeline).

The research results of the influence of emitter power on the destruction of GH granules of a given concentration and size are shown in Fig. 5.

Analysis of the obtained results shows that for the destruction of small particles (<0.01 mm) even if their concentration is low (0.01–0.02%), a relatively high-power emitter (3–5 kW) is required. The same is observed in the case of GH accumulation to high concentrations (>0.1%) with a particle size of 0.05–0.1 mm. Therefore, the most advantageous strategy for combating GH accumulation is its active destruction when it reaches sizes of 0.02–0.05 mm, which do not harm the course of the main technological process. The emitter power should be sufficient to maintain the GH concentration within acceptable limits. In particular, with an input concentration of seed crystals up to 0.05%, a magnetron with a power of 1 kW is sufficient.

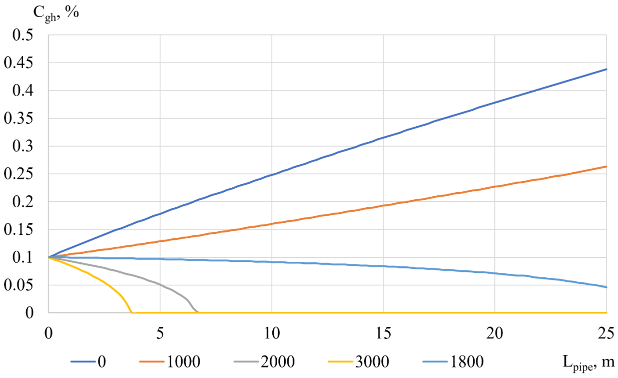

Fig. 4. Change in GH concentration along the length of the gas pipeline at different EM emitter power: 0, 1000, 1800, 2000, 3000 W

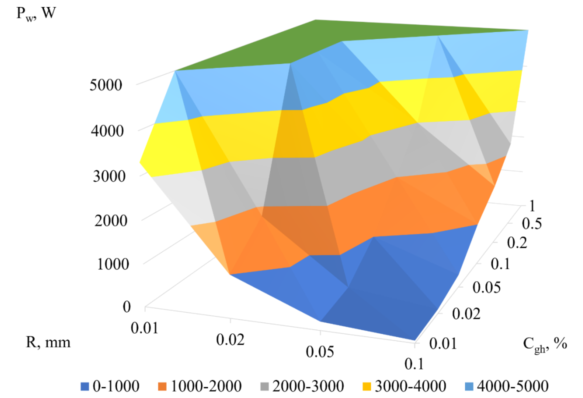

Fig. 5. Required emitter power for the destruction of GH particles of initial size (R) and initial concentration (C_{gh}) (in the legend – EM emitter power, W)

Statistical analysis of the results of mathematical modeling allows to propose the following approximation dependence for determining the minimum emitter power, which, given the given initial data, leads to a decrease in the gas hydrate concentration in the gas pipeline, W

$$P_w = 124C_{gh} / R^{1.7}, \tag{28}$$

where C_{gh} – the initial gas hydrate concentration, %; R is the size of gas hydrate particles, mm. The obtained dependence allows to quickly determine the required emitter power in engineering calculations.

3.3. Discussion

The proposed model combines two processes of synthesis and dissociation of gas hydrates, taking into account the hydrodynamic processes that occur in the pipeline. The developed mathematical model is similar to that described in [12] in considering heat and mass transfer processes on a spherical surface, however, in this work the processes were considered on the surface of a water drop. Also, the general principles of absorption of microwave EM radiation by gas hydrate, applied in the mathematical model, coincide with those set forth in works [8–13], however, the types of waves and the state of the medium in which they are absorbed differ.

The main differences of the proposed mathematical model from the existing ones are:

1. Simultaneous consideration of two mutually opposing processes of synthesis and dissociation of gas hydrate on the surface of the granule.
2. These processes occur during the movement of the gas hydrate granule inside the gas pipeline.
3. Heating of the gas hydrate granule occurs as a result of its absorption of multimode microwave EM direct and reflected radiation.

Factors that limit (reduce the accuracy) of the use of the mathematical model are: the induction period of gas hydrate formation in the model is considered complete, although in reality it is difficult to predict the exact time of the appearance of seed crystals; the variable nature of the multiphase medium in the gas pipeline; lack of consideration of agglomeration of gas hydrate particles; change in the salt composition of water can shift the thermodynamic equilibrium, which is not taken into account in the mathematical model.

Possible directions for improving the mathematical model are to take into account the polydispersity of gas hydrate particles. It is also worth developing a laboratory setup for a comprehensive research of the heat and mass transfer processes of synthesis/dissociation of gas hydrate granules inside the gas pipeline under the influence of microwave EM radiation.

According to the results of mathematical modeling, a complex approximation dependence was obtained for determining the required emitter power $Pw = 124C_{gh}/R^{1.7}$, W, which allows calculating the required minimum magnetron power to combat hydrate formation in the pipeline.

The obtained dependences are the basis for improving gas preparation technologies in conditions of hydrate formation by using microwave radiation energy.

Further scientific research can be aimed at increasing the energy efficiency of the method and conducting experimental tests on laboratory stands to confirm theoretical calculations. In addition, the adaptation of the technology to pipelines of different diameters is promising, which will allow the method to be scaled up and implemented as a reliable alternative to traditional chemical methods of combating hydrates.

4. Conclusions

1. A mathematical model has been proposed, which, unlike existing ones, simultaneously takes into account the dynamics of two opposite processes: the kinetics of the synthesis of gas hydrate microgranules in the flow and their dissociation under the influence of microwave radiation energy. The model is based on the heat balance of the interfacial surface of the granule, which allows for a high-accuracy prediction of the critical radius of the particle at which its growth stops.
2. It has been established that the use of microwave electromagnetic radiation allows changing the regime of "avalanche" growth of hydrate concentration to the regime of their stabilization or complete destruction. It has been proven that the efficiency of dissociation depends on the selective absorption of energy: small particles (R = 0.01 mm) require a higher specific radiation power for destruction due to intensive heat removal into the cooled gas medium, which justifies the need to maintain optimal power parameters to prevent particle coalescence.
3. Based on the results of multimode modeling (modes H_{11}, E_{01}, H_{01}), an analytical dependence $P_W = 124C_{gh}/R^{1.7}$ was obtained, which allows determining the required emitter power depending on the current concentration of hydrates and the size of their granules. This creates a methodological basis for automating the control of ultrahigh-frequency installations in low-temperature gas preparation systems in order to minimize the use of chemical inhibitors.

Conflict of interest

The authors declare that they have no conflict of interest regarding this research, including financial, personal, authorship or other nature, which could affect the research and its results presented in this paper.

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Data availability

Data cannot be provided for the reasons specified in the data availability statement.

Use of artificial intelligence

In this work, the authors used GPT-5.2 for grammatical, linguistic and stylistic correction. The authors manually checked and edited the material and confirm that artificial intelligence tools were only auxiliary and were not used to generate and formulate the aim, objectives, methodology, analyze the results, or formulate conclusions.

Authors' contributions

Taras Podoliak: Software, Formal analysis, Writing – original draft, Writing – review and editing, Visualization; ***Viktoriia Dmytrenko***: Conceptualization, Methodology, Investigation.

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