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MATHEMATICAL MODEL OF HYDRATE FORMATION IN THE FLOW
OF MOIST GAS IN TUBES

V. M. Bilyushov

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The problem of hydrate formation in the flow of moist gas in tubes is formulated, under the assumption that the temperature of hydrate formation depends not only on the pressure but also on the water-vapor concentration at the phase-transition surface.

The problem of hydrate formation in gas pipelines was first considered in [1-3], where the conditions of hydrate formation were described, and recommendations for the prevention of hydrate formation in gas-pipeline operations were made; these reduce to the need to dry the gas, remove condensed water, and employ pipeline operating conditions that eliminate the possibility of hydrate formation. The problem was then discussed in [4-10], where attempts were made to determine in advance the sites of possible obstruction of the pipeline by hydrates, and to give a quantitative calculation of the mass of hydrate forming in the course of gas transport. However, these works have a series of deficiencies. In [8], for example, the mass rate of hydrate formation was estimated, but no mention was made of which section of the pipeline was subject to hydrate deposition. In [9], the region of possible hydrate formation was determined on the basis of the thermodynamic conditions of moisture removal from the gas, but the process of hydrate deposition itself was not considered. In [10], the model of hydrate formation was constructed from the numerical solution of the equations of nonisothermal motion of a real gas, and the action of the hydrate obstruction was modeled by a local resistance with an unknown drag coefficient, which is a significant deficiency of the model.

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In all these works, it is assumed that the process of hydrate-layer growth at the pipeline walls is determined solely by the conditions of precipitation of the moisture. No account is taken here of heat transfer between the moving gas and the hydrate, and the influence of the hydrate layer on the gas flow is not considered, i.e., no account is taken of the influence of the change in through cross section of the tube on the flow parameters, which means that it is impossible to consider the dynamics of hydrate-layer growth over time.

In [11], the problem of hydrate-layer growth in a pipeline was solved within the framework of the Stefan problem, i.e., it was assumed that the growth rate of the hydrate layer is determined by heat-transfer processes between the gas flow and the hydrate layer. It was assumed that the free moisture required for hydrate formation is sufficient over the whole length of the tube. In real conditions, all the moisture in the pipeline is in the vapor state.

It is known that the formation of hydrate requires the satisfaction of two conditions simultaneously: thermodynamic and moisture-content conditions. Below, a mathematical model of hydrate formation in the flow of a real moist gas in a tube is proposed, taking account of both these conditions.

The mathematical model is based on the following physical model: the process of hydrate formation begins at the tube wall, since the gas close to the wall is considerably supercooled in comparison with the flow core and its velocity of motion is a minimum, which facilitates the retention of newly formed hydrate at the tube wall. It is assumed that the rate of hydrate-layer growth is determined both by the heat transfer from the gas to the tube wall and by the supply of moisture to the reaction surface. In this framework, the process of hydrate formation and growth in gas flow in tubes must be described by a conjugate problem of heat and mass transfer.

Since gas flow in boreholes and gas pipelines has a clearly expressed turbulent character, the equations describing the gas flow in the tube become one-dimensional, and take the form [12]

$$\begin{aligned}
 & \frac{\partial}{\partial t} (\rho S_w) + \frac{\partial}{\partial x} (\rho V S_w) = \Pi_w \rho_w (D_n - V_n)_w, \\
 & \frac{\partial}{\partial t} (\rho V S_w) + \frac{\partial}{\partial x} [(P + \rho V^2) S_w] = P \frac{\partial S_w}{\partial x} + \Pi_w \left(\mu \frac{\partial U}{\partial r} \right)_w - \rho g S_w \sin \gamma, \\
 & \frac{\partial}{\partial t} \left[\rho S_w \left(i + \frac{V^2}{2} \right) \right] + \frac{\partial}{\partial x} \left[\rho V S_w \left(i + \frac{V^2}{2} \right) \right] = \frac{\partial}{\partial t} (P S_w) + \\
 & + \frac{\partial}{\partial x} \left(\lambda S_w \frac{\partial T}{\partial x} \right) - \rho g V S_w \sin \gamma + \Pi_w \left(\lambda \frac{\partial T}{\partial r} \right)_w + \Pi_w \rho_w i_h (D_n - V_n)_w, \\
 & \frac{\partial}{\partial t} (\rho S_w \Theta) + \frac{\partial}{\partial x} (\rho V S_w \Theta) = \Pi_w [\rho \Theta (D_n - V_n) + j_n]_w.
 \end{aligned} \tag{1}$$

Assuming that the change in thickness of the hydrate layer over the tube length is small, and using the condition of "adhesion" at the hydrate-layer boundary, it is found that

$$D_{nw} = \frac{\partial R_w}{\partial t}, \quad V_{nw} = V_w, \quad j_{nw} = \rho_w \left(D \frac{\partial C}{\partial r} \right)_w. \tag{2}$$

The density at the phase-transition boundary is assumed to be the same as in the flow core, i.e., $\rho_w = \rho$.

At the phase-transition boundary, there is a discontinuous change in a series of parameters. Taking account of the immobility of the hydrate, the absence of moisture flow in the hydrate, and Eq. (2), the conditions at the discontinuity surface are written in the form

$$\Pi_w \rho_w (\dot{R}_w - V_w) = \Pi_w \rho_h \dot{R}_w,$$

$$\Pi_w \left(\lambda_h \frac{\partial T_h}{\partial r} \right)_w - \Pi_w \left(\lambda \frac{\partial T}{\partial r} \right)_w = \Pi_w \rho_h L \dot{R}_w, \quad (3)$$

$$\Pi_w \rho_h \dot{R}_w (\Theta_w - \Theta_h) = - \Pi_w \rho_w \left(D \frac{\partial C}{\partial r} \right)_w$$

Using Eqs. (2) and (3), the system in Eq. (1) may be brought to the form

$$\begin{aligned} \frac{\partial}{\partial t} (\rho S_w) + \frac{\partial}{\partial x} (\rho V S_w) &= \rho_h S_w, \quad S_w = \frac{\partial \dot{S}_w}{\partial t}, \\ \frac{\partial}{\partial t} (\rho V S_w) + \frac{\partial}{\partial x} [(P + \rho V^2) S_w] &= P \frac{\partial S_w}{\partial x} - \rho g S_w \sin \gamma + \Pi_w \left(\mu \frac{\partial U}{\partial r} \right)_w, \\ \frac{\partial}{\partial t} \left[\rho S_w \left(i + \frac{V^2}{2} \right) \right] + \frac{\partial}{\partial x} \left[\rho V S_w \left(i + \frac{V^2}{2} \right) \right] &= \frac{\partial}{\partial t} (P S_w) + \\ + \frac{\partial}{\partial x} \left(\lambda S_w \frac{\partial T}{\partial x} \right) - \rho g V S_w \sin \gamma + \Pi_w \left(\lambda \frac{\partial T}{\partial r} \right)_w + \rho_h i_h S_w, \\ \frac{\partial}{\partial t} (\rho S_w \Theta) + \frac{\partial}{\partial x} (\rho V S_w \Theta) &= \rho_h \Theta_h S_w. \end{aligned} \quad (4)$$

The relation between the pressure, temperature, and moisture concentration at the gas-hydrate interface is determined by the equation of state of the vapor-gas mixture, which may be written in the form

$$\Theta_w = \frac{a(P, T_w)}{\frac{P}{P_{H_2O}^h} - 1 + a(P, T_w)}, \quad a = \frac{\mu_{H_2O}}{\mu_g} \frac{z_g(P, T_w)}{z_{H_2O}(P, T_w)}. \quad (5)$$

There are a number of formulas for calculating the water vapor pressure above the hydrate. Thus, for dry methane gases in the temperature range from 0 to -50°C , the recommended equation in [5]

$$P_{H_2O}^h = \exp(24.87 - 6235/T - 0.1593 \ln P). \quad (6)$$

In [13], the following equation was proposed

$$\lg(P_{H_2O}^h/y) = -V_{H_2O}(P-y)/RT. \quad (7)$$

The water vapor pressure above the hydrate may also be calculated by means of the Barrer-Stuyart equation

$$P_{H_2O}^h = P_{H_2O}^0 \prod_{m=1,2} \left(1 + \sum_j C_{mj} P_j \right)^{-K_m}, \quad (8)$$

where $P_{H_2O}^0$ is determined [14] from the equation

$$\lg P_{H_2O}^0 = A - \frac{B}{T} - C \lg T. \quad (9)$$

The system in Eq. (4) is complemented by the equation of state of the gas mixture in the flow core

$$P = \rho RT \left[\frac{z_{H_2O}}{\mu_{H_2O}} \Theta + \frac{z_g}{\mu_g} (1 - \Theta) \right] \quad (10)$$

and the thermodynamic relation

$$di = C_p dT + \left[v - T \left(\frac{\partial v}{\partial T} \right)_p \right] dP. \quad (11)$$

The heat and mass transfer will be assumed to conform to the Newton and Fick laws, while the tangential stress is determined from the Darcy-Weisbach law

$$\begin{aligned} \left(\lambda \frac{\partial T}{\partial r} \right)_w &= \alpha (T_w - T), & \left(D \frac{\partial C}{\partial r} \right)_w &= \beta (\Theta_w - \Theta), \\ - \left(\mu \frac{\partial U}{\partial r} \right)_w &= \frac{\psi}{8} \rho V^2. \end{aligned} \quad (12)$$

In determining the heat- and mass-transfer coefficients, a two-layer model of turbulent transfer is taken as the starting point, and it is assumed that there is a region between the core of the gas flow and the hydrate layer in which the temperature and concentration change from T and Θ in the flow core to T_w and Θ_w at the phase-transition surface. In this transfer scheme, the heat- and mass-transfer coefficients may be written in the form

$$\alpha = \lambda/\delta, \quad \beta = D/\delta, \quad (13)$$

where δ is the thickness of the region between the flow core and the surface of the hydrate layer, equal to the thickness of the laminar sublayer

$$\delta = 11.5 \frac{v}{u_*}, \quad u_* = \sqrt{\frac{\psi}{8}} V. \quad (14)$$

Using Eq. (12) and assuming a quasisteady temperature distribution in the hydrate layer, it follows from Eq. (3) that

$$\dot{R}_w = \frac{\lambda_h (T_w - T_N)}{L \rho_h R_w \ln R_w/R_0} - \frac{\alpha (T_w - T)}{L \rho_h}, \quad (15)$$

$$\dot{R}_w = \frac{\beta \rho}{\Theta_h \rho_h} (\Theta_w - \Theta). \quad (16)$$

If the pipeline is laid in the ground, the heat transfer between the pipeline and the ground must be taken into account. In this case, the thermal condition in Eq. (15) takes the form [15]

$$\dot{R}_w = \frac{T_w - T_N}{L \rho_h R_w \left(\frac{1}{\lambda_h} \ln R_w/R_0 - \frac{1}{\lambda_{gr} G} \right)} - \frac{\alpha (T_w - T)}{L \rho_h}, \quad (15')$$

where G is the thermal resistance of the perturbed ground

$$G = \max \left\{ \frac{1}{\alpha_0}; \frac{R-1}{R \ln R - R + 1} \right\}, \quad \alpha_0 = \ln \left(\frac{H}{R_0} + \sqrt{\frac{H^2}{R_0^2} - 1} \right).$$

The radius of perturbation of the ground $R(t)$ is determined by solving an ordinary differential equation [15].

From Eqs. (15) and (16), an equation for the unknown concentration at the phase interface is obtained

$$\Theta_w = \Theta + \frac{\Theta_h}{\beta \rho L V S_w} \left[\frac{2 \sqrt{\pi} \lambda_h (T_w - T_N)}{\ln S_w/S_0} - \alpha \sqrt{S_w} (T_w - T) \right]. \quad (17)$$

Using Eq. (5), Θ_w is eliminated from consideration, and an equation is obtained for determining the temperature at the phase-transition boundary

$$\begin{aligned} \Theta + \frac{\Theta_h}{\beta \rho (P, T) L V S_w} \left[\frac{2 \sqrt{\pi} \lambda_h (T_w - T_N)}{\ln S_w/S_0} - \right. \\ \left. - \alpha \sqrt{S_w} (T_w - T) \right] = \frac{a(P, T_w)}{\frac{P}{P_{H_2O}^h(P, T_w)} - 1 + a(P, T_w)}. \end{aligned} \quad (18)$$

Thus, Eqs. (4), (10), (11), (15), and (18) form a closed system of equations for the variables ρ , V , S_w , P , T , i , Θ , T_w .

Note that, excluding from consideration the last equation of the system in Eqs. (8) and (9), and assuming that the phase-transition temperature depends only on the pressure, according to the law $\ln P = A - B/T_w$, the formulation of the problem in [11] is obtained.

Quasisteady conditions of gas flow in the pipeline are considered. It is necessary to make a series of assumptions here, simplifying the basic system of equations: that the change in thickness of the hydrate layer over time and along the tube is sufficiently small that the mass flow rate of gas is much larger than the mass growth rate of the hydrate; that the heat conduction in the axial direction may be neglected, for both the gas and the hydrate; that the physical properties of the gas and the hydrate are constant; that the pressure, temperature, and concentration at the inlet are constant; that the change in moisture concentration along the tube axis is determined by the conditions of its transition to the hydrate state.

Taking account of the assumptions adopted, the system in Eq. (4) takes the form

$$\rho V S_w = M = \text{const}, \quad \frac{dP}{dx} = \frac{\Delta_1}{\Delta}, \quad \frac{dT}{dx} = \frac{\Delta_2}{\Delta}, \quad \frac{d\Theta}{dx} = \frac{\rho_h \Theta_h}{M} \frac{dS_w}{dt}. \quad (19)$$

The determinants Δ , Δ_1 , and Δ_2 are as follows

$$\Delta = \begin{vmatrix} 1 + \frac{M^2}{S_w^2} \left(\frac{\partial v}{\partial P} \right)_T; & \frac{M^2}{S_w^2} \left(\frac{\partial v}{\partial T} \right)_P \\ -T \left(\frac{\partial v}{\partial T} \right)_P; & C_p \end{vmatrix},$$

$$\Delta_1 = \begin{vmatrix} \frac{M^2 v}{S_w^3} \frac{dS_w}{dx} - \frac{g \sin \gamma}{v} - \frac{V \sqrt{\pi} M^2 v \psi}{4 S_w^{2.5}}; & \frac{M^2}{S_w^2} \left(\frac{\partial v}{\partial T} \right)_P \\ \frac{2\alpha \sqrt{\pi} S_w (T_w - T)}{M} + \frac{V \sqrt{\pi} M^2 v^2 \psi}{4 S_w^{2.5}}; & C_p \end{vmatrix},$$

$$\Delta_2 = \begin{vmatrix} 1 + \frac{M^2}{S_w^2} \left(\frac{\partial v}{\partial P} \right)_T; & \frac{M^2 v}{S_w^3} \frac{dS_w}{dx} - \frac{g \sin \gamma}{v} - \frac{V \sqrt{\pi} M^2 v \psi}{4 S_w^{2.5}} \\ -T \left(\frac{\partial v}{\partial T} \right)_P; & \frac{2\alpha \sqrt{\pi} S_w (T_w - T)}{M} + \frac{V \sqrt{\pi} M^2 v^2 \psi}{4 S_w^{2.5}} \end{vmatrix}$$

Then Eq. (15) is rewritten in the form

$$\frac{dS_w}{dt} = \frac{4\pi\lambda_h(T_w - T_N)}{L\rho_h \ln S_w/S_0} - \frac{2\alpha \sqrt{\pi} S_w (T_w - T)}{L\rho_h}. \quad (20)$$

The calculation algorithm is constructed as follows.

1. With a fixed cross section $S_w(x)$ over time, $T_w(0)$ is found from the specified values P_0 , T_0 , and Θ_0 in the inlet cross section of the tube by means of Eq. (18).

2. Using the result obtained for $T_w(0)$, the system in Eq. (19) is integrated over one step in x .

3. From the values of $P(\Delta x)$, $T(\Delta x)$, $\Theta(\Delta x)$ found at the end of the integration interval, $T_w(\Delta x)$ is determined, and integration over the next interval is performed, and so on, as long as $x \leq l$, the length of the pipeline.

4. Making a time step, a new value of the cross-sectional area is obtained from Eq. (20), and the calculation procedure is repeated.

This algorithm may also be used in calculating the process of hydrate formation in a borehole or gas pipeline that is not in use. Even in such a borehole, the gas is not at rest, because the cap at the end of the borehole leaks a certain amount of the gas. The mass flow rate of gas in unused boreholes amounts to 5000 m³/day, but even at flow rates of ~ 1000 m³/day the flow in the borehole is turbulent, since Re ~ 7 · 10³ in this case. Therefore, the above formulation of the problem is valid in the present case. Numerical calculation allows the safe standing time of a borehole to be determined.

For a short tube, the change in the parameters over the length may be neglected. In the limiting case, a system of equations describing the growth of the hydrate layer in a fixed tube cross section may be obtained from Eq. (4).

$$\begin{aligned} \frac{dS_w}{dt} &= \frac{4\pi\lambda_h(T_w - T_N)}{L\rho_h \ln S_w/S_0} - \frac{2\alpha V\sqrt{\pi S_w}(T_w - T)}{L\rho_h}, \quad S_w(0) = S_0, \\ \frac{dV}{dt} &= -g \sin \gamma - \frac{\rho_h V}{\rho S_w} \frac{dS_w}{dt} - \frac{\psi}{4} \sqrt{\frac{\pi}{S_w}} V^2, \quad V(0) = V_0, \\ \frac{d\Theta}{dt} &= \frac{\rho_h(\Theta_h - \Theta)}{\rho S_w} \frac{dS_w}{dt}, \quad \Theta(0) = \Theta_0, \\ \frac{dP}{dt} &= \frac{\Delta'_1}{\Delta'}, \quad P(0) = P_0, \quad \frac{dT}{dt} = \frac{\Delta'_2}{\Delta'}, \quad T(0) = T_0, \end{aligned} \quad (21)$$

where

$$\begin{aligned} \Delta' &= \rho S_w^2 \cdot \left[C_P \left(\frac{\partial \rho}{\partial P} \right)_T + T \left(\frac{\partial \rho}{\partial T} \right)_P \left(\frac{\partial v}{\partial T} \right)_P \right], \\ \Delta'_1 &= \rho S_w C_P (\rho_h - \rho) \frac{dS_w}{dt} - S_w \left(\frac{\partial \rho}{\partial T} \right)_P \cdot \left[(P - \rho_h L) \frac{dS_w}{dt} - \rho V S_w \left(\frac{dV}{dt} + g \sin \gamma \right) + 2\alpha \sqrt{\pi S_w} (T_w - T) \right], \\ \Delta'_2 &= S_w \left(\frac{\partial \rho}{\partial P} \right)_T \cdot \left[(P - \rho_h L) \frac{dS_w}{dt} - \rho V S_w \left(\frac{dV}{dt} + g \sin \gamma \right) + 2\alpha \sqrt{\pi S_w} (T_w - T) \right] + \rho S_w T (\rho_h - \rho) \left(\frac{\partial v}{\partial T} \right)_P \frac{dS_w}{dt}. \end{aligned}$$

A case that is important for practical applications is now considered: gas flow in a tube in the presence of local resistances. The latter significantly disrupt the conditions of gas flow in their immediate vicinity. At local resistances, there is a loss in pressure head, which is associated with pressure variation, in the general case. As a result of the throttling of the gas, its temperature rises, and hence there is a discontinuity in its density and velocity. On account of the change in flow parameters of the gas, accelerated precipitation of hydrates occurs close to the local resistances. The following algorithm is proposed for the calculation of the hydrate formation with gas flow in a tube with local resistances.

Using the system in Eq. (19), the calculation is performed in the section of smooth tube up to the first local resistance. From the solution of this system, the flow parameters preceding the local resistance (denoted by the subscript "minus") are found. Then, the parameters following the resistance (subscript "plus") are determined from the results for the parameters preceding it, by means of the system of equations

$$\begin{aligned} \rho_+ V_+ S_+ &= \rho_- V_- S_- = M, \\ \Theta_+ &= \Theta_-, \\ P_+ &= P_- - \xi \frac{\rho_- V_-^2}{2}, \\ T_+ &= T_- - \varepsilon(P_-, T_-)(P_- - P_+), \\ \rho_+ &= \rho_+(P_+, T_+). \end{aligned} \quad (22)$$

These results for the parameters are then taken as the initial values for the calculation in the section of tube up to the next resistance.

NOTATION

V , ρ , P , i , mean (over the tube cross section) velocity, density, pressure, and enthalpy; $v = 1/\rho$, specific volume of gas; Θ , Θ_h , moisture concentration in the gas flow and in the hydrate; R_w , Π_w , S_w , radius, perimeter, and cross-sectional area of the gas flow; T , C , V_w , U , local values of the temperature, concentration, and the normal and tangential components of the velocity vector; D_n , velocity of phase-interface motion in the direction of its normal; j_n , normal component of the flow of material at the phase-transition surface; ν , μ , λ , D , kinematic and dynamic viscosity, heat conduction, and diffusion coefficient; t , x , r , time and spatial coordinates; L , latent heat of phase transition; y , $P_{H_2O}^o$, $P_{H_2O}^h$, saturated water vapor pressure above water, an empty hydrate lattice, and hydrate; V_{H_2O} , molar volume of liquid water; C_{11} , Langmuir constants; α , β , ψ , heat- and mass-transfer coefficients and hydraulic drag coefficient; l , local drag coefficient; H , depth at which gas pipeline is laid; R_o , S_o , radius and cross-sectional area of tube; R , radius of thermal perturbation of ground; z , compressibility; ϵ , Joule-Thompson coefficient; μ_{H_2O} , μ_g , molecular weight of water and gas. Indices: w , h , g , H_2O , gr , phase interface, hydrate, gas, water, and ground.

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