Journal of Engineering Physics and Thermophysics, Vol. 78, No. 3, 2005

HEAT AND MASS TRANSFER IN CHEMICAL TRANSFORMATIONS

EXPERIMENTAL INVESTIGATION OF THE DECOMPOSITION OF A NONPOLAR-GAS HYDRATE IN A PIPE UNDER THE ACTION OF A MICROWAVE ELECTROMAGNETIC FIELD

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Results of experimental investigations of the decomposition of a propane hydrate in a pipe under the action of a microwave electromagnetic field are presented. Methods of obtaining a gas hydrate, its decomposition under the action of a microwave electromagnetic field, and measuring the temperature and pressure in a pipeline have been developed. It has been established that the rate of decomposition of a gas hydrate increases under the action of a microwave electromagnetic field. It is shown that a gas hydrate in a pipe can be completely decomposed under the action of a microwave electromagnetic radiation whose wavelength is comparable to the thickness of this hydrate.

In the process of production and transportation of gas and oil, paraffin and gas hydrates are formed in different parts of pipelines [1, 2]. This decreases the carrying capacity of pipelines and can lead to their breakage. Therefore, the removal of paraffin and gas hydrates from pipelines is an important problem in oil and gas production.

At present, there is no universal method of removal of paraffin and gas hydrates from oil and gas wells and pipelines. This is explained, first of all, by the fact that the mechanisms of formation of paraffin and gas hydrates in the process of production of oil and gas are different and different (in design) wells and pipelines are used for their production and transportation. In practice, the following methods are used for removal of plugs from pipelines and wells: the pressure in a pipeline or well is decreased at a constant temperature; the temperature in a pipeline is increased to a value exceeding the temperature of decomposition of a gas hydrate or the temperature of separation of paraffin; substances decomposing plugs are introduced into a well.

Paraffin and gas hydrates represent dielectrics with a complex relative permittivity [3]:

$$\dot{\varepsilon}(\omega, T, P) = [\varepsilon'(\omega, T, p) - j\varepsilon''(\omega, T, p)] , \quad j = \sqrt{-1} . \tag{1}$$

The imaginary part of the dielectric constant determines, as is known, the density of heat sources that are formed in a material under the action of a high-frequency electromagnetic field:

$$q = 0.5\omega\varepsilon'\varepsilon_0 \tan \delta E^2 , \quad \tan \delta \approx \frac{\varepsilon''}{\varepsilon'} . \tag{2}$$

The internal heat sources arising in such a dielectric under the action of a high-frequency electromagnetic field cause changes in its temperature and pressure, which is used for decomposition of paraffin- and gas-hydrate plugs formed in different units of equipment used for production of oil and gas [4, 5]. One of the advantages of this method is liberation of additional heat in the equipment material having a limiting conductivity [5].

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 1062-0125/05/7803-0524

It should be noted that effects and processes arising in different materials under the action of electromagnetic fields are being extensively investigated for the purpose of development of new technological processes [4–13] and methods of decomposition and melting of various deposits in equipment used for production and transportation of oil and gas.

Tubing strings, gushing tubes, and gas and oil pipelines represent, from the electrodynamics standpoint, a circular waveguide filled with an inhomogeneous dielectric. An electromagnetic wave with a wavelength smaller than the critical wavelength will propagate in such a waveguide with attenuation [14, 15]. It was proposed in [9, 10] to use this effect for removal of viscous and solid deposits, possessing dielectric losses, from pipelines. Under the action of such a wave, solid deposits are melted and viscous deposits lose their viscosity. In this way, a pipeline can be cleared from the undesirable substances.

The possibility of removal of an oil plug from a long pipeline as a result of its heating by a high-frequency electromagnetic radiation was investigated in [12]. It was shown in this work that a 7-km plug of a high-paraffin oil in a pipe (the diameter of the pipe was not involved) can be decomposed over the course of several days at a rate of 40–60 m/h under the action of an electromagnetic radiation of power 30–50 kW. Judging from the materials of the indicated work, in it, an ideal case, where all electromagnetic energy supplied from an oscillator was used for heating and melting of the paraffin plug, was considered. The critical frequencies at which electromagnetic waves effectively propagate in a pipe and the losses in its metal walls were not taken into account.

The possibility of prevention of the formation of gas hydrates in pipes and removal of such hydrates with the use of a microwave electromagnetic field was experimentally investigated in [11]. A pipe of diameter 76 mm and length 16 m was used in the experiments. Prior to the electromagnetic action, a gas flow was throttled through the pipe under pressure for 50–55 min from 18–20^{o}C to 9–11^{o}C, with the result that the pressure in it increased to 5.3 MPa. This pointed to the formation of a gas hydrate in the pipe. To decompose the gas hydrate formed the pipe was subjected to the action of a pulsed electromagnetic wave of power 90 kW (mean power 60 W). Under the action of this wave, the gas temperature increased and, as a result, the gas hydrate decomposed, which was evidenced by the decrease in the gas pressure in the pipe to the initial value, detected after approximately 1 h.

In [16], the power and time of action of an electromagnetic wave with a damping coefficient of ~0.1 m^{-1} that are necessary for removal of a plug in a pipeline have been estimated. It has been shown that, at a radiation power of ~10 kW, the time of heating of a 100-m plug in a pipe of diameter 0.1 m is equal to 100 h. In this case, the radiation frequency should be equal to 2.5–3.0 GHz, i.e., it should be higher than the allowed industrial frequencies.

A gas hydrate in a pipe heated by microwave radiation breaks down into gas and water. In the case where a gas hydrate is heated intensively and the gas-water mixture formed as a result of its decomposition is removed rapidly, a disperse flow arises. In a vertical pipe, two regions are formed in this case. One of these regions is occupied by the gas-water mixture and the other region is occupied by the solid hydrate located under the gas-water mixture. The resonance properties of the gas-water mixture representing a waveguide, in which electromagnetic waves are excited, have been theoretically investigated. It has been established that the density and disposition of heat sources in the mixture are determined by its dimensions. This conforms with the theory of propagation of electromagnetic waves in stratified media [17]. As a result of the interaction of an incident wave with the wave reflected from the wave phase boundary, a standing electromagnetic wave arises in the first region.

In [5], the influence of a microwave electromagnetic field on the melting of paraffin in a short-circuited coaxial system has been investigated. The dynamics of melting of paraffin and resins and decomposition of hydrates in a long coaxial line, in the interelectrode space of which an electromagnetic field of frequency 2400 MHz is generated, was investigated with the use of an experimental setup whose schematic diagram was presented. It has been shown that the temperature distribution along the depth of a well is inhomogeneous and corresponds to the distribution of the standing electromagnetic wave field in an interelectrode space, and the presence of paraffin in it significantly accelerates the phase transition in the whole coaxial system.

Certain features of the decomposition of a gas hydrate in the equipment used for production of gas and oil have been experimentally revealed in [18–20]. However, the mechanism of decomposition of a gas hydrate remains not clearly understood. This is explained, in part, by the lack of experimental data on the indicated process.

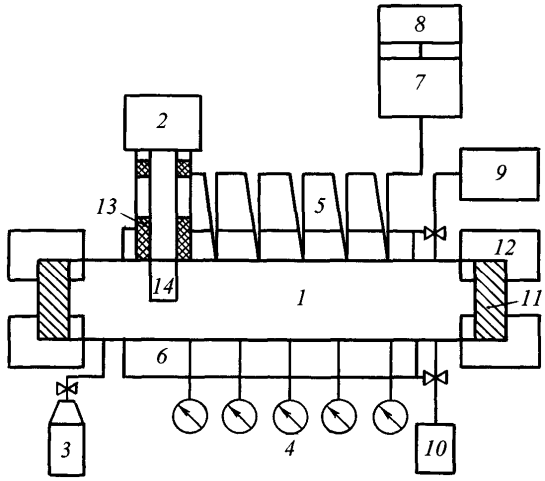


Fig. 1. Block diagram of the experimental setup.

The aim of the present work is to experimentally investigate the mechanism of decomposition of a nonpolar-gas (propane) hydrate [3] in a pipe under the action of a microwave electromagnetic field.

Description of the Experimental Setup. The mechanism of decomposition of a gas hydrate in a pipe under the action of a microwave electromagnetic field was experimentally investigated on a setup, the block diagram of which is presented in Fig. 1. It includes a model of a pipeline, a microwave oscillator, a coaxial line, a hydraulic system, control-measuring instruments, and a cooling system.

The microwave electromagnetic energy is supplied to pipeline 1 through emitter 14, a copper post connected to the rectangular waveguide of oscillator 2 with the use of a coaxial-waveguide connector. Emitter 14 is 3.2 cm (a quarter of the length of the electromagnetic wave in the air) high above the inner surface of the pipe.

A Parus oscillator was used in the experiments. It has the following characteristics: rated microwave power, not less than 2.2 kW; range of continuous adjustment, 0–20 dB; range of change in the standing-wave ratio in the dynamic regime, 1.5–10; oscillation frequency, 2375 MHz $\pm$ 1%; maximum required power, 6.5 kW; power supply from a three-phase power network, 380/220 V or 220/127 V; industrial- or potable-water discharge, 7–10 liters/min; weight, 280 kg.

A gas hydrate in a pipeline represents, from the electrodynamics standpoint, a circular guide; therefore, it can break down if a certain-configuration electromagnetic field is excited in it. Waves of H_{11} type mainly propagate in a circular waveguide [14, 15] because their losses are minimum as compared with the losses of the other waves. This allows the H_{11} waves to be used for microwave decomposition of a large-length gas hydrate in a pipeline.

The length of a working H_{11} wave in a waveguide of radius a should satisfy the condition $\lambda < \lambda_{cr}$. An electromagnetic wave can propagate in a circular waveguide only in the case where the operating frequency of an electromagnetic-energy source satisfies the condition $f > f_{cr}$ or $\lambda < \lambda_{cr}$ [14]. In our experiment, f = 2375 MHz. According to the condition $\lambda/2.61 > a > \lambda/3.14$, an H_{11} wave can be excited in a pipeline of radius $0.036 \text{ m} < a < 0.048 \text{ m}$.

The radius of the pipeline forming a part of the setup developed by us is 4 cm, which meets the above-mentioned condition. The critical frequency of an H_{11} wave in this pipeline is determined by the formula [14]

$$f_{cr} = \frac{1.841}{2\pi \cdot 0.04} \frac{1}{\sqrt{\varepsilon_0 \mu_0}} \approx 2.2 \text{ HGr} .$$

Consequently, the condition $f > f_{cr}$ is fulfilled.

To create conditions under which the temperature and pressure along the pipeline could be measured simultaneously, we used a waveguide of length 50.4 cm comprising the four lengths of an electromagnetic wave in the air and estimated the amount of gas necessary for filling the pipeline. Since the ends of the waveguide are transparent for microwave electromagnetic waves, it is necessary that the radiation loss be minimum or absent, i.e.,

$$\frac{P(z)}{P_0} = \exp(-2\alpha z) . \tag{3}$$

The absorption coefficient α of a microwave electromagnetic wave in a pipe with a gas-hydrate filler was calculated by the formula $\alpha = \alpha_m + \alpha_d$, where [14, 15]

$$\alpha_m = \sqrt{\frac{\pi f \mu \mu_0}{\sigma}} \frac{1}{Z_{f.m} a \sqrt{1 - \left(\frac{\lambda_0}{\lambda_{cr}}\right)^2}} ; \quad \alpha_d = \frac{\pi \varepsilon' \tan \delta}{\lambda_0 \sqrt{1 - \frac{1}{\varepsilon'} \left(\frac{\lambda_0}{\lambda_{cr}}\right)^2}} . \tag{4}$$

At f = 2375 MHz, $\sigma = 0.34 \cdot 10^7\ \Omega^{-1} \cdot m^{-1}$, μ = 2.72 H/m [21], $Z_{f.m} = \sqrt{\mu_0 / \varepsilon_0} \approx 377\ \Omega$, a = 0.04 m, f_{cr} = 2.2 GHz, ν_{11} = 1.841, ε' = 3.75, and $\tan \delta$ = 0.02 [3], from formulas (4) we obtain $\alpha_m = 0.02\ m^{-1}$, $\alpha_d = 2.13\ m^{-1}$, and $\alpha = 2.15\ m^{-1}$. Consequently, the electromagnetic-energy losses in a pipe depend substantially on the amount of gas hydrate in it. Under the above-indicated conditions, the electromagnetic-field energy does not penetrate through the pipe to the environment, which is very important for experiments in which the application of microwave energy is studied.

The hydraulic system of the setup includes a cylinder filled with gas 3 and a pump for supply of water 10. The cylinder and pump are connected to the pipeline through the corresponding valves (see Fig. 1). The cooling system comprises thermal jacket 6 connected to a thermostat. The control-measuring instruments are manometers 4 and copper-constantan thermocouples 5. Unions for monomers and thermocouples are welded on the side surface of the cylinder at a distance of 6 cm. The ends of the thermocouples are run through the unions in the pipeline at the level of its inner surface. The thermocouples are connected to an M95 millivoltmeter 7 through distributor 8. The thermocouples are made of constantan and copper magnet wires of cross section 0.2 mm^2. The thermocouples were graduated in advance. A definite temperature was maintained in the pipeline with the use of thermal jacket 6, which was connected to the thermostat by unions. The ends of the pipeline were made from acrylic plastic 11 and encapsulated with covers 12, which is necessary for visual observation of the formation and decomposition of gas hydrates.

The working volume of the cylinder was equal to 30 dm^3. Fluoroplastic sealant 13 was used for sealing the site of introduction of a microwave electromagnetic wave. The internal and external electrodes of the coaxial line were separated by fluoroplastic washers. The amount of gas introduced into the pipeline was measured by flowmeter 9.

Experimental Procedure. The experimental procedure was as follows:

(1) determination of the working capacity of the experimental setup;

(2) investigation of the thermohydrodynamic effects arising in the process of melting of ice under the action of a microwave field;

(3) perfection of the method of obtaining a gas hydrate in a pipeline;

(4) investigation of the mechanism of decomposition of a gas hydrate in a pipeline under the action of a microwave electromagnetic field.

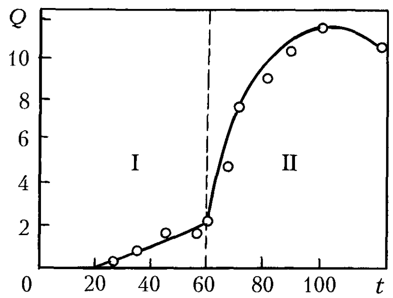
We have performed ten series of such experiments.

The pressure tests have shown that the setup considered can work at pressures as high as 9.5–10.0 MPa.

To determine the influence of a microwave electromagnetic field on the rate of melting of ice, we determined the amount of water flowing from the pipeline in the case where a microwave electromagnetic action was absent, i.e., at room temperature (22^{o}C). In our experiments, water appeared at the output of the pipe after 17–18 min from the beginning of the experiment. As is seen from Fig. 2 (region 1), in the absence of microwave action, the rate of water flow at the output of the pipe did not exceed 2–3 ml/min. Thereafter the pipeline was exposed to a microwave electromagnetic field in accordance with the block diagram in Fig. 1. The microwave power supplied to the pipeline was calculated by the formula

$$\Delta P = P_{w.t} - P_t = 0.07 CV (T_2 - T_3) .$$

The measurements have shown that T_2 = 17.1^{o}C and T_3 = 12^{o}C. Therefore, the electromagnetic-field power supplied from the microwave oscillator is equal to $P = 257 \pm 1$ W. It is seen from Fig. 2 that the amount of water



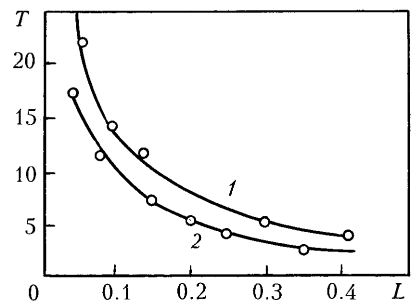
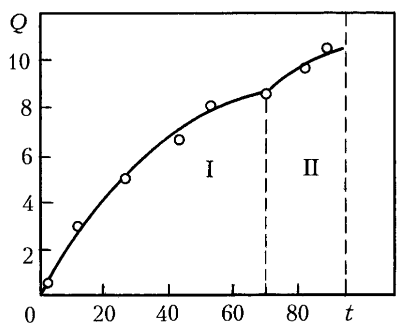


Fig. 2. The rate of ice melting at room temperature (I) and under the action of a microwave electromagnetic field (II).

Fig. 3. Temperature distribution along the model pipeline: 1) gas hydrate; 2) ice; t = 90 min.



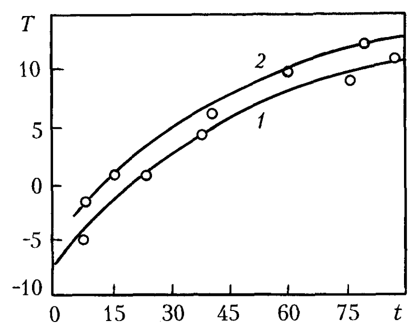


Fig. 4. Amount of gas flowing from the model pipeline: I) method of decreasing pressure; II) method of microwave action.

Fig. 5. Change in temperature in the model pipe line under the action of a microwave electromagnetic field: 1) ice; 2) gas hydrate.

flowing from the pipeline increases sharply when the pipe is exposed to a microwave electromagnetic field, which points to the fact that the rate of ice melting increases under the action of this field. In this case, the rate of water flow at the output of the pipeline reaches 12 dm^3/min. This points to the fact that the rate of ice melting increases by 5–6 times as compared with the rate of its natural melting. Then, when the microwave electromagnetic field continues to act on the pipeline, the amount of water flowing from it somewhat decreases (Fig. 3, curve 2), which is explained by the nonlinear temperature distribution along the pipeline.

Figure 3 shows the temperature distribution along the pipeline considered. The temperature at the emitter is higher than the temperature at the output of the pipeline, i.e., it decreases with distance from the emitter, which is in qualitative agreement with dependence (3). Thus, it may be concluded that the setup developed by us is capable of working, is safe, and can be used for investigating the thermodynamic effects arising in the process of melting of ice and a gas hydrate under the action of a microwave field.

Results of Experiments on Decomposition of a Gas Hydrate. We investigated the decomposition of a propane hydrate in the model pipeline under the action of a microwave field. A gas hydrate was produced in the following way. Half the pipeline volume was filled with distilled water, which was then frozen in a refrigerating chamber. Thereafter the ice was partially defrosted and propane was pumped into the pipeline until the pressure was 0.45 MPa. Then the pipeline was placed in the refrigerating chamber once again and held at a temperature from -2^oC to $+1^oC$. The gas hydrate was formed over the course of 5–6 days. The process was controlled visually through the end windows. As the known diagram, presented in [22], shows, a gas hydrate is formed or decomposed in two cases: as a result of an increase in the pressure or a decrease in the temperature of the gas in a pipeline. We first attempted to decompose a gas hydrate by decreasing the pressure in the pipeline. The amount of gas flowing from the model pipeline (measured by a flowmeter) increased nonlinearly and reached a constant value with time (Fig. 4). Then the pipeline was exposed to a microwave electromagnetic field with simultaneous measurement of the amount of gas flowing

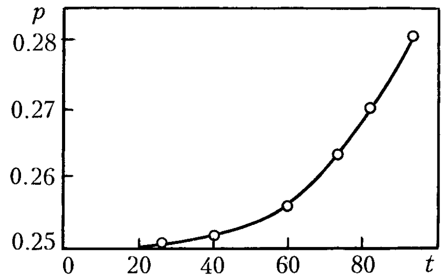


Fig. 6. Change in pressure in the model pipeline in the case of decomposition of a gas hydrate under the action of a microwave electromagnetic field.

from the pipeline. As is seen from Fig. 4 (region II), the amount of gas flowing from the pipeline increases sharply. Thus, a microwave electromagnetic action increases the efficiency of the method of decreasing the pressure in a pipeline.

Of interest are the results of investigation of the decomposition of a gas hydrate only under the action of a microwave field at a constant pressure, i.e., without removal of gas. It is seen from Fig. 5 (curve 2) and Fig. 3 (curve 1) that a gas hydrate is heated more rapidly than ice. The gas hydrate is heated more strongly than ice because the values of ε' and tan δ are larger for it than for ice [3]. In our experiments, the gas hydrate in the pipeline was completely decomposed for 84 min; this time is much shorter than the time of its natural decomposition at room temperature. The pressure in the pipeline was increased by 0.03 MPa for 30 min (Fig. 6), which is explained by the fact that the temperature in the pipeline was increased as a result of the microwave action (Fig. 3) in accordance with the Gay–Lussac law.

CONCLUSIONS

1. The thermodynamic effect arising in the process of action of a microwave electromagnetic field on a gas hydrate and leading to its decomposition has been detected. It has been established that the rate of decomposition of a gas hydrate is much larger than that of ice, which is explained by the difference in their dielectric parameters.

2. The depth of effective action of an electromagnetic microwave on a propane hydrate in a pipe is approximately equal to the length of the acting wave in this pipe, representing a circular waveguide. This makes it possible to effectively use a microwave electromagnetic field for decomposition of small-length gas-hydrate plugs in units of equipment used for production of gas and oil.

3. The energy necessary for decomposition of one cubic meter of a gas hydrate in a pipe by the action of a microwave electromagnetic field is equal to approximately 2000 kW·h.

4. The experimental setup developed by us and the data obtained with the use of this setup can be used for substantiation of the technology of decomposition of gas hydrates in pipelines and paraffin hydrates in gas and oil wells with the use of a microwave electromagnetic field.

NOTATION

a, radius of the pipe, m; *C*, heat capacity of water, J/(kg·K); *E*, electric-field strength, V/m; *f*, electromagnetic-wave frequency, Hz; H_{11}, main type of electromagnetic waves propagating in a circular waveguide; *L*, length of the model pipeline, m; *p*, pressure, MPa; $P(z)$, power of the electromagnetic field at cross section *z* of the pipeline, W; P_0, power of the electromagnetic field at the point of introduction of the microwave-field energy into the pipeline, W; $P_{\mathrm{w.t}}$, power of the electromagnetic field in the waveguide of a microwave oscillator without connection to the experimental setup, W; P_{t}, power of the electromagnetic field in the waveguide of a microwave oscillator connected to the experimental setup, W; *Q* amount of gas flowing from the pipeline, dm^3; *q*, density of volume heat sources; *T*, temperature, $^{\mathrm{o}}$C; T_2 and T_3, temperatures of the outflowing water in the case where the pipeline is not connected to the waveguide and in the case where it is connected to the waveguide, $^{\mathrm{o}}$C; tan δ, dielectric loss tangent; *t*, time, min; *V*, volumetric rate of the air flow, m^3/sec; *z*, cylindrical coordinate, m; Z_{m}, wave resistance, Ω; α, coefficient of absorption of a microwave electromagnetic wave, m^{-1}; ε' and ε'', real and imaginary parts of the dielectric constant of the medium; ε_0, permittivity of free space, F/m; λ, length of an electromagnetic wave, m; λ_0, length of an electromagnetic wave in a vacuum, m; μ_0, permeability of free space, H/m; μ, relative permeability; ν_{11}, first root of the equation $J_1'(x) = 0$, where $J_1(x)$ is the Bessel function of the first kind; σ, electrical conduction of the pipe material, $\Omega^{-1}\cdot m^{-1}$; $\omega = 2\pi f$, cyclic frequency, rad/sec. Subscripts: d, dielectric; cr, critical; m, material of the pipe; t, transmission line; w.t, without transmission line; f.m, filling material.

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