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MOTION OF GAS-LIQUID SYSTEMS, TAKING ACCOUNT OF MICRONUCLEUS FORMATION
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A model is proposed for the motion of a gas-liquid system in tubes. The theoretical and experimental resul.ts are in good agreement.

The complexity and multiplicity of the flow systems leads to considerable difficulties in studying the motion of gas-liquid systems in tubes, both in conducting the experiments and in constructing the mathematical models. Nevertheless, there have already been many experimental and theoretical investigations of the motion of gas-liquid flows in tubes (see [1-5], etc.), in which processes at pressures below the saturation pressure are mainly considered.

However, gas-1iquid flows at pressures above or close to the saturation pressure are investigated as homogeneous systems, as a rule, on the assumption that transitions from one state to another occur instantaneously in the equilibrium thermodynamic theory of phase transitions.

However, according to the data of [6], the formation of a new phase occurs not instantaneously over the whole volume but rather takes the form of local fluctuations passing beyond the limits of a single aggregate state. Nuclei of new phase (gas bubbles) are "heterophase" and it is assumed, in accordance with the results of [6], that in the region above and especial. ly close to the saturation pressure the system is not completely homogeneous. The "heterophase" system may be both in equilibrium and in a nonequilibrium state. As a rule, in the given conditions, the dispersed gas is uniformly distributed over the liquid volume.

Experiment shows [7, 8] that in a point volume the pressure level and its rate of change influence the formation of micronuclei. In connection with this, experiments are conducted to determine the moment of appearance of micronuclei of the gas phase. In a container connected to a press, a gasified liquid is prepared; it consists of transformer oil and carbon dioxide at the saturation pressure ( 0.04 MPa ). Then the pressure is increased systematically to 0.25 MPa , i.e., considerably above the saturation pressure. Then the pressure drops systematically reduced at a definite rate to different levels above the saturation pressure. Analysis of the experimental results shows that, beginning at some value ( $\mathrm{P}=0.17 \mathrm{MPa}$ ), the pressure increases over time. As the pressure approaches the saturation level, it increases more rapidly (Fig. 1); this may be due to the formation of micronuclei of gas phase.

The presence of micronuclei in the system leads to a significant dependence of the density on the pressure and the rate of change in pressure, and probably is responsible for the decrease in values of the rheological parameters observed experimentally in. [9, 10].

Taking account of the foregoing, a model may be proposed for the motion of gas-liquid systems in tubes at pressures above or close to the saturation pressure.

1. The system of differential equations for the motion of the gas-liquid medium is written as in [11]

[^0]

Fig. 1. Pressure variation over time at various levels above the saturation pressure. P.10, MPa; t, sec.

$$
\begin{gather*}
\frac{\partial\left(f_{\rho} W\right)}{\partial t}+\frac{\partial\left(f \rho W^{2}\right)}{\partial x}=-f \frac{\partial P}{\partial x}+\chi \tau \\
\frac{\partial(f \rho)}{\partial t}+\frac{\partial(f \rho W)}{\partial x}=0 \tag{1}
\end{gather*}
$$

After some manipulations, Eq. (1) may be written in the form

$$
\begin{gather*}
\frac{\partial W}{\partial t}+\frac{1}{2} \frac{\partial W^{2}}{\partial x}=-\frac{1}{\rho_{0}} \frac{\partial P}{\partial x}+\frac{\chi}{\rho_{0} f_{0}} \tau(W)  \tag{2}\\
\frac{\partial(f \rho)}{\partial t}=-\frac{\partial(\rho f W)}{\partial x}
\end{gather*}
$$

Suppose that the cross-sectional area of the tube depends on the pressure according to the law

$$
\begin{equation*}
f=f_{0}\left(1+a \frac{P-P_{0}}{E}\right) \tag{3}
\end{equation*}
$$

Since it is assumed that the formation of new phase is not instantaneous, a relation between the density and pressure may be used [12]

$$
\begin{equation*}
\frac{\rho_{0}}{k_{\mathrm{c}}^{0}}\left(P-P_{0}+\theta_{\rho} \frac{d P}{d t}\right)=\rho-\rho_{0}+\frac{k_{\mathrm{c}}^{\infty}}{k_{\mathrm{c}}^{0}} \theta_{\rho} \frac{d \rho}{d t} \tag{4}
\end{equation*}
$$

The dependence of the stress on the velocity is specified as for a viscous liquid [11]

$$
\begin{equation*}
\tau=-\frac{4 \mu}{R} W \tag{5}
\end{equation*}
$$

In the presence of gas inclusions distributed over the whole volume of the liquid, the viscosity of the system may be written analogously [13]

$$
\begin{equation*}
\mu=\mu_{0}(1+\gamma C) \tag{6}
\end{equation*}
$$

The value of $\gamma$ may be both positive (in [13], a value of 2.5 was adopted for spherical solids) and negative (for gas inclusions).

Probably, Eq. (6) may be used for the motion of a gas-liquid system and, at a pressure below the saturation pressure, to describe bubble conditions.

The concentration value depends on the pressure level and its rate of change, and also on the disequilibrium of the inclusions and the solution of the gas phase, which allows the following phenomenological relation to be written

$$
\begin{equation*}
\theta \frac{d C}{d t}+C=\beta\left[\left(P-P_{\mathrm{s}}\right)+\lambda \frac{d P}{d t}\right] \tag{7}
\end{equation*}
$$

or

$$
\begin{equation*}
C=\beta\left(P-P_{s}\right)-\frac{\beta(\theta-\lambda)}{\theta}\left[P_{(0)}-P_{\mathrm{s}} \mathrm{e}^{-\frac{t}{\theta}}-\frac{\beta(\theta-\lambda)}{\theta} \int_{0}^{t} \mathrm{e}^{-\frac{t-t_{1}}{\theta}} \frac{d P}{d t_{1}} d t_{1}\right. \tag{8}
\end{equation*}
$$

Since processes at pressures $P \leq P_{s}$ are considered, $\beta<0$.
Together with Eqs. (3)-(8), Eq. (2) forms a complete system of equations of isothermal motion of a gasified liquid in tubes at pressures above or close to the saturation pressure. Substituting Eqs. (3)-(8) into Eq. (2), it is found after some adjustments (neglecting the nonlinear terms in the continuity equation) that

$$
\begin{gather*}
-\frac{1}{\rho_{0}} \frac{\partial P}{\partial x}-\frac{1}{2} \frac{\partial W^{2}}{\partial x}=\frac{\partial W}{\partial t}+2 a\left[1+\beta \gamma\left(P-P_{s}\right)+\right. \\
\left.+\frac{\beta \gamma(\theta-\lambda)}{\theta}\left[P_{(0)}-P_{\mathrm{s}}\right] \mathrm{e}^{-\frac{t}{\theta}}-\frac{\beta \gamma(\theta-\lambda)}{\theta} \int_{0}^{t} \mathrm{e}^{-\frac{t-t_{1}}{\theta}} \frac{d P}{d t_{1}} d t_{1}\right] W  \tag{9}\\
\frac{c_{\infty}^{2}}{c_{0}^{2}} \theta_{\rho}\left[\frac{c^{2}}{c_{\infty}^{2}}\left(1-\frac{c_{\infty}^{2}}{c_{0}^{2}}\right)+1\right] \frac{\partial^{2} P}{\partial t^{2}}+\frac{\partial P}{\partial t}=-\rho_{0} c^{2}\left(\frac{\partial W}{\partial x}+\frac{c_{\infty}^{2}}{c_{0}^{2}} \theta_{\rho} \frac{\partial^{2} W}{\partial t \partial x}\right),
\end{gather*}
$$

where $2 a=4 \mu_{0} \chi / R \rho_{0} f_{0}=8 \mu_{0} / \rho_{0} R_{0}^{2}\left(f_{0} / \chi=R / 2 \quad\right.$ for a circular tube $) ; c^{2}=k_{\mathrm{c}}^{0} / \rho_{0}\left(1+e \frac{k_{\mathrm{c}}^{0}}{E}\right) ; c_{\infty}^{2}=k_{\mathrm{c}}^{\infty} / \rho_{0} ; \quad c_{0}^{2}=$ $k_{\mathrm{c}}^{0} / \rho_{0} ; e=d / \delta$.

The nonlinear terms in the continuity equation are estimated as follows. In

$$
\frac{\partial f \rho W}{\partial x}=\rho \frac{\partial W f}{\partial x}+W f \frac{\partial \rho}{\partial x}=\rho \frac{\partial W f}{\partial x}+\frac{W f}{c^{2}} \frac{\partial P}{\partial x}
$$

the first term $\rho \frac{\partial W f}{\partial x}$ is of order $\rho_{0} f_{0} W / L$ and the second $\sim f_{0} W_{0} P / c^{2} L$. The nonlinear term may be neglected if $f_{0} \rho_{0} W / L 》>f_{0} W P / c^{2} L$, i.e., $P / c^{2} \rho_{0} \ll 1$.

For liquids of density $\rho_{0} \sim 10^{3} \mathrm{~kg} / \mathrm{m}^{3}$ at a pressure of the order of 1.0 MPa and a velocity of propagation of the perturbations of $10^{3} \mathrm{~m} / \mathrm{sec}$, the latter conditions is satisfied with sufficient accuracy ( $10^{-3} \ll 1$ ).
2. Consider the steady motion of a gas-liquid system at a pressure no lower than the saturation pressure. It is assumed that

$$
\frac{\partial W}{\partial t}=0, \quad \frac{\partial P}{\partial t}=0, \quad \frac{\partial^{2} P}{\partial t^{2}}=0, \quad \frac{\partial W}{\partial t \partial x}=0, \quad \frac{\partial W^{2}}{\partial x}=0, \quad P_{(0)}=P_{s}
$$

Then it follows from the second relation in Eq. (9) that $W=$ const. The first relation of this system is

$$
\begin{equation*}
-\frac{1}{\rho_{0}} \frac{\partial P}{\partial x}=2 a\left[1+\beta \gamma\left(P-P_{s}\right)-\frac{\beta \gamma(\theta-\lambda)}{\theta} \int_{0}^{t} \mathrm{e}^{-\frac{t-t_{1}}{\theta}} \frac{d P}{d t_{1}} d t_{1}\right] W . \tag{10}
\end{equation*}
$$

Taking account of the expression for the total derivative

$$
\frac{d P}{d t_{1}}=\frac{\partial P}{\partial t_{1}}+\frac{d x}{d t_{1}} \frac{\partial P}{\partial x}=\frac{\partial P}{\partial t_{1}}+W \frac{\partial P}{\partial x}
$$

and the stationarity condition $\partial \mathrm{P} / \partial \mathrm{t}_{1}=0$, Eq. (10) is written in the form

$$
\begin{equation*}
-\frac{1}{\rho_{0}} \frac{\partial P}{\partial x}=2 a\left[1+\beta \gamma\left(P-P_{\mathrm{s}}\right)-\frac{\beta \gamma(\theta-\lambda)}{\theta} W \int_{0}^{t} \mathrm{e}^{-\frac{t-t_{1}}{\theta}} \frac{d P}{d x} d t_{1}\right] W . \tag{11}
\end{equation*}
$$

Taking account of the expression $d / d t=W d / d x$, Eq. (11) is differentiated with respect to $t$, to give

$$
\begin{equation*}
-\frac{1}{\rho_{0}} W \frac{d^{2} P}{d x^{2}}=2 a \beta \gamma\left(W \frac{d P}{d x}+\frac{\theta-\lambda}{\theta^{2}} W \int_{0}^{t} \mathrm{e}^{-\frac{t-t_{1}}{\theta}} \frac{d P}{d x} d t_{1}-\frac{\theta-\lambda}{\theta} W \frac{d P}{d x}\right) W . \tag{12}
\end{equation*}
$$

After some transformations, using Eq. (11), Eq. (12) is written in the form

$$
\begin{equation*}
\theta \frac{d^{2} P}{d x^{2}}+\left(2 a \beta \gamma \lambda \rho_{0} W+\frac{1}{W}\right) \frac{d P}{d x}+2 a \beta \gamma \rho_{0} P=2 \alpha \gamma \beta \rho_{0} P_{\mathbf{s}}-2 \alpha \rho_{0} . \tag{13}
\end{equation*}
$$

The following condition is assumed in the initial cross section

$$
\begin{equation*}
P(0)=P^{0} ; \quad \frac{d P(0)}{d x}=2 a \rho_{0} W . \tag{14}
\end{equation*}
$$

The solution of Eq. (13) with the condition in Eq. (14) takes the form

$$
\begin{gather*}
P=P_{\mathrm{s}}-\frac{1}{\gamma \beta}-\frac{1}{k_{1}-k_{2}}\left[\left(P^{0}-P_{\mathrm{s}}+\frac{1}{\gamma \beta}\right)\left(k_{2} \mathrm{e}^{k_{1} x}-k_{1} \mathrm{e}^{k_{2} x}\right)+2 a \rho_{0} W\left(\mathrm{e}^{k_{1} x}-\mathrm{e}^{k_{2} x}\right)\right], \\
k_{1,2}=\frac{-\left(2 a \gamma \beta \lambda \rho W+\frac{1}{W}\right) \pm \sqrt{\left(2 a \gamma \beta \lambda \rho W+\frac{1}{W}\right)^{2}-4 \cdot 2 a \gamma \beta \rho \theta}}{2 \theta} . \tag{15}
\end{gather*}
$$

If $k_{i} Z \ll 1$, which is characteristic for short pipes and large times of micronucleus formation, Eq. (15) is brought to the following form by appropriate manipulations

$$
\begin{equation*}
\Delta P=2 a \rho_{0} W l\left(1-2 a \gamma \rho_{0} W l \frac{\lambda}{2 \theta}\right)+\left(P^{0}-P_{\mathrm{s}}\right) \frac{2 a \gamma \beta \rho_{0} l^{2}}{2 \theta} . \tag{16}
\end{equation*}
$$

It is evident from Eq. (16) that the presence of micronuclei of new gas phase in the system leads to reduction in pressure drop; this effect decreases with increase in $\theta$. If $k_{i} l \gg 1$, however, which is characteristic for long pipes and infinitesimal times of micronucleus formation, it follows from Eq. (15) that

$$
\begin{equation*}
\Delta P=2 a \rho_{0} W l-\frac{1}{\gamma \beta}+P_{\mathrm{s}}-P_{l} . \tag{17}
\end{equation*}
$$

Thus, pressure losses in gas-liquid systems depend on the pressure $P_{S}$ and the coefficients characterizing the presence of gas inclusions in the system; since the pressure loss in the gas-liquid system is less than in a homogeneous medium, the condition $1 / \gamma \beta+P_{\mathcal{I}}>P_{s}$ must be satisfied.
3. To elucidate the features of the motion of a gas-liquid system in tubes at pressures above and close to the saturation pressure and test the adequacy of the chosen model, experiments are performed on the apparatus shown in Fig. 2. Its basic elements are the mixer 1, the high-pressure cylinder 2, a differential manometer 3 , and tube 4 of length 1.3 m .


Fig. 2. Schematic diagram of experimental apparatus.


Fig. 3. Dependence of the flow rate of the gas-liquid system on the pressure difference with various rates of pressure change: 1,2 ) reduction and increase in mean pressure level, respective1 y . $\mathrm{Q}, \mathrm{cm}^{3} / \mathrm{sec} ; ~ \mathrm{P} \cdot 10^{3}, \mathrm{MPa}$.

A gas-1iquid mixture consisting of transformer oil and carbon dioxide is prepared in the mixer. The volume of liquid phase $V_{L}=25 \mathrm{dm}^{3}$, the gas volume in normal conditions $V_{G}=7.5$ $\mathrm{dm}^{3}$. The saturation pressure is maintained at $\mathrm{P}_{\mathrm{S}}=0.03 \mathrm{MPa}$ and the temperature of the system is $293^{\circ} \mathrm{K}$.

The system is held for 60 min to reach the saturation pressure. Then the pressure in the mixture is increased to $P=0.15 \mathrm{MPa}$ (without mixing) and the measurements are made of the mixture discharging with change in pressure difference, which is regulated by the degree to which the valve (cutoff valve) at the tube outlet is opened. The liquid volume is first measured with increase in pressure difference; the dependence $Q(\Delta P)$ is represented by a straight line here, as for viscous incompressible liquid.

On reaching the maximum pressure difference under the condition that the pressure at the tube outlet is larger than the saturation pressure, the dependence $Q(\Delta P)$ is determined with decrease in pressure difference to complete equalization. In this case, the dependence $Q(\Delta P)$ is rectilinear up to some value of the pressure difference, and then (at a differences of the order of $9.2-10.5 \cdot 10^{-3} \mathrm{MPa}$ ) deviates to a lower straight line, and an anomaly appears (hysteresis) (Fig. 3).

The experimental data are described using Eq. (10). In the experiments, the rate of pressure variation is constant, i.e., $\mathrm{dP} / \mathrm{dt}=\mathrm{A}=$ const. Taking account of the foregoing and the condition of stationarity of the experiments, Eq. (10) is written in the form

$$
\begin{equation*}
-\frac{1}{\rho_{0}} \frac{d P}{d x}=2 a\left[1+\beta \gamma\left(P-P_{\mathrm{s}}\right)-A \beta \gamma(\theta-\lambda)\right] W \tag{18}
\end{equation*}
$$

or

$$
\begin{equation*}
\frac{d P}{d x}+2 a \rho_{0} \beta \gamma W P=-2 a \rho_{0} W\left[1-\beta \gamma P_{s}-A \beta \gamma(\theta-\lambda)\right] \tag{19}
\end{equation*}
$$

The solution of Eq. (19) under the condition $P(0)=P^{0}$ takes the form

$$
\begin{equation*}
P=\left\{P^{0}+\frac{1}{\beta \gamma}\left[1-\beta \gamma P_{\mathrm{s}}-A \beta \gamma(\theta-\lambda)\right]\right\} \mathrm{e}^{-2 a \rho_{0} \beta \gamma W{ }^{W} x}-\frac{1}{\beta \gamma}\left[1-\beta \gamma P_{\mathrm{s}}-A \beta \gamma(\theta-\lambda)\right] . \tag{20}
\end{equation*}
$$

Since $P=P_{\ell}$ when $x=\eta$, taking into account that $2 a_{\rho_{0} \beta \gamma} W l \ll 1$ in laboratory conditions, simple manipulations reduce Eq. (20) to the form

$$
\begin{equation*}
\Delta P=P^{0}-P_{l}=2 a \rho_{0} W l\left[1+\beta \gamma\left(P^{0}-P_{\mathrm{s}}\right)-A \gamma \beta(\theta-\lambda)\right] . \tag{21}
\end{equation*}
$$

Analysis of Eq. (21) shows that the pressure difference depends not only on the gas content but also on the ratio of pressure change in the system; with reduction in pressure ( $\mathrm{A}<0$ ), the pressure difference is somewhat larger than with increase in pressure in the system (A>0).

The experimental results in Fig. 3 are analyzed according to Eq. (21) with the following parameter values: $\mu=16.9 \mathrm{~Pa} \cdot \mathrm{sec} ; \rho=880 \mathrm{~kg} / \mathrm{m}^{3} ; Z=1.3 \mathrm{~m} ; \mathrm{P}^{\circ}=0.15 \mathrm{MPa} ; \mathrm{P}_{\mathrm{S}}=0.16 \mathrm{MPa}$

$$
\left|\frac{d P}{d t_{1}}\right|=|A|=2,85 \cdot 10 \mathrm{MPa} / \mathrm{sec}
$$

The calculations give the result $2 a \rho_{0} W=5,51 \cdot 10^{-2} \quad(\mathrm{MPa} / \mathrm{m}) \mathrm{sec}$.
If

$$
\begin{aligned}
& A<0, \quad \frac{\Delta P}{W}=5,5 \cdot 10^{-2}(\mathrm{MPa} / \mathrm{m}) \mathrm{sec} \\
& A>0, \quad \frac{\Delta P}{W}=3,25 \cdot 10^{-2}(\mathrm{MPa} / \mathrm{m}) \mathrm{sec}
\end{aligned}
$$

Using the values obtained for $2 a \rho_{0} W$ and the two values of $\Delta P / W$, Eq. (21) gives: $\beta \gamma=2$. $10^{-1} 1 / \mathrm{MPa} \theta-\lambda=7.2 \cdot 10^{3} \mathrm{sec}$.

The theoretical straight lines plotted from Eq. (21) are shown as dashed lines in Fig. 3 and are in adequate agreement with experimental data. Some discrepancy in the region where the rate of pressure change passes from plus to minus may be explained in that the system was held at each pressure level for an insufficient period in the experiments.

## NOTATION

$f$, cross-sectional area of tube; $\rho$, liquid density; $W$, mean liquid velocity over the cross section; $P$, pressure; $\bar{X}$, wetted perimeter; $\tau$, tangential stress; $x$, direction of flow; $t$, time; $f_{0}$, cross-sectional area of tube at pressure $P_{0} ; d$, coefficient depending on the form of the cross section and the wall thickness; E, elastic modulus of tube material; $\mathrm{k}_{\mathrm{c}}^{\circ}$, $k_{c}^{\infty}$, bulk-compression modulus of system in slow and fast processes, respectively; $\rho 0$, density at pressure $P_{0} ; \theta_{\rho}$, relaxation time of system; $\mu$, viscosity of system; R, tube radius; $\mu_{0}$, liquid viscosity; $C$, concentration of gas inclusions in liquid; $P_{S}$, pressure at which the first gas micronuclei are formed in the liquid; $\theta$, $\lambda$, times characterizing the formation of micronuclei and the rate of pressure variation.

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## MARAGONI CONVECTION IN A TWO-LAYER SYSTEM

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Equations are obtained that govern the characteristics of weakly nonlinear concentration and thermocapillary convection stabilized by the nonlinearity of convective transport of impurity mass and heat.

Under unstable surface tension conditions depend on the impurity concentration or temperature on the interface of two immiscible fluids, the appearance of the instability of the mass or heat transport process through this boundary (Marangoni effect) is possible. For plane interfacial surfaces such an instability was apparently first considered in. [1-3], and was then investigated in a very large number of papers with diverse physicochemical factors and the curvature of the surface itself taken into account.

Ordered or chaotic convective motions (interphasal convection or turbulence) which are capable of significant intensification of the mass as well as heat transport through this surface [4-6], are formed as a result of the mentioned instability in domains adjacent to the interfacial surface. Thus, for instance, the effect mass transfer coefficient during extraction in liquid-1iquid systems can be increased because of natural or artificially produced Marangoni convection by 2-10 times as compared with analogous stable systems [7].

If known numerical investigations of interphasal convective structures of the type performed in [8] are excluded from consideration, then attempts at a nonlinear analysis of the result of the appearance of the Marangoni instability, which are perfectly necessary for estimation of the parameters of these structures, are limited, in practice, to weakly nonlinear problems to which the method of the small parameter developed in [9-11] is applicable. However even in this case the calculations and final equations turn out to be quite awkward, which requires their asymptotic and numerical analysis for comparatively simple particular situation.
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