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The law governing the size distribution of detached gas-liquid streams of drops has been determined analytically, and a comparison is carried out against experimental data existing in the literature. The derived theoretical relationships offer an excellent description of existing experimental results.

The detachment of drops from the surface of a fluid streamlined by a stream of gas (vapor) is observed in steam separators, in nozzles, in the operating grids of turbines, as well as in a variety of atomization devices. In all of these cases one of the most important parameters of the process is the size of the forming drop, and in order to achieve correct calculations it is important not only to know the average values but also the law governing drop distribution. At the present time, based on an experimental study of drop formation this is assumed to be a normal law or one that is logarithmically normal [1, 2]. However, it seems to us that such an approach is not fully justified. First of all, the combination of a group of drops into one of uniform size is arbitrary to a considerable extent and, secondly, the postulated normal law nevertheless requires empirical information regarding the average size and dispersion that occurs in each specific case, which makes the theoretical description of the process extraneous.

In the present study we undertake an analytical derivation of the law of drop distribution by size on fluid detachment from wave crests at a film surface attracted by a stream of gas. In this case, the process of drop formation exhibits the following characteristics [3]: the surface of the film is covered by waves whose crests periodically detach and are entrained in the form of drops exhibiting various diameters. The detachment occurs from large waves in a turbulent gas core. Between the large waves there exists a minor ripple that does not participate in the drop formation.

Let us examine the liquid particle participating in the detachment process. This particle is affected by the force of inertia m'a, balanced by the detaching force  $F_0$  and the force of adhesion  $F_{ad}$ :

$$m'a = F_0 - F_{\rm ad}.$$
 (1)

At the instant of separation, the force of adhesion  $F_{ad} = \sigma \ell$  assumes a zero value. If we assume that the velocities of streamlining are sufficiently large and that the force of separation is determined primarily by the magnitude of the relative velocity, it may be assumed to be constant in the period from the onset of detachment all the way to its conclusion. It then follows from (1) that

$$F_0 = m'a_0. \tag{2}$$

The onset of separation is characterized by equality between the forces of separation and adhesion, i.e., zero acceleration, so that with consideration of (2) we therefore derive from (1) the following relationship:

$$m'a_0 = \sigma L. \tag{3}$$

The law of motion for a solitary drop allows us to write the following expression for the left-hand side of equality (3):

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$$m'a_0 = cs\rho'' - \frac{u_0^2}{2}.$$
 (4)

The coefficient c here, according to [1], is determined from a formula of the form

$$c = k_1 \left(\frac{2u_0 r_0}{v''}\right)^{k_s},\tag{5}$$

where  $k_1$  and  $k_2$  depend on Re =  $2u_0r_0'/v''$ .

From (3) and (4) we obtain

$$s^{-\frac{1}{2}} = \frac{u_0}{[2\sigma L (c\rho^{11})^{-1}]^{1/2}},$$
 (6)

or with consideration of the sphericity of the separating drops (s =  $\pi r_0^{12}$ )

$$(r'_0)^{-1} = \frac{u_0}{[2\sigma L (\pi c \rho'')^{-1}]^{1/2}}.$$
(7)

Let us introduce the notation:

$$X_{1} = \frac{u_{0}R}{v''}; \quad X_{2} = \left(\frac{2\sigma L}{\pi c \,\rho'' \,v''^{2}}\right)^{1/2}; \quad X_{3} = \frac{R}{r_{0}'}.$$

Then equality (7) is written in the form

$$X_3 = \frac{X_1}{X_2}.$$
(8)

Let us examine the random quantities  $X_1$ ,  $X_2$ ,  $X_3$ . We will assume these to be constant, having the following density distributions  $f_1(x_1)$ ,  $f_2(x_2)$ ,  $f_3(x_3)$ , respectively. If  $f(x_1; x_2)$  is the density of distribution for the vector  $(X_1; X_2)$ , then we can write the relationship from [4] as follows:

$$f_{\mathbf{s}}(x_{\mathbf{s}}) = \int_{0}^{\infty} \alpha f(\alpha x_{\mathbf{s}}, \alpha) d\alpha - \int_{-\infty}^{0} \alpha f(\alpha x_{\mathbf{s}}, \alpha) d\alpha,$$

and in the case of the independence and nonnegativeness of  $X_1$  and  $X_2$ 

$$f_3(x_3) = \int_0^\infty \alpha f_1(\alpha x_3) f_2(\alpha) d\alpha.$$
(9)

Let us return once again to the random quantity  $X_1 = u_0'' - u_0'$ . It represents the difference in the velocities of the gas at the point of separation and the drop that has just been formed. For each lateral coordinate z this difference randomly assumes some value from the range defined, for example, by the intensity of the turbulent pulsations of the flow. We can then regard the probability of the event  $F_1(x_1) = P_1(X < x_1)$  to be dependent on the parameter z, while the totality of events  $X_1 < x_1(z)$  can be treated as a space of elementary events. Correspondingly, the total derivative of the function  $F_1$  with respect to the parameter z is equal to 0, as a result of which for  $f_1(x_1) = \partial F_1/\partial x_1$  the following continuity equation is valid:

$$\frac{dF_1}{dz} = \int_0^{x_1 \max} \left[ \frac{\partial f_1}{\partial z} + \frac{\partial}{\partial z} \left( \frac{\partial x_1}{\partial z} f_1 \right) \right] dx_1 = 0.$$
(10)

The velocity gradient along the lateral coordinate depends exclusively on the transverse coordinate and therefore  $dx_1/dz = \varphi(z)$ . In this case, any function of the following form will serve as a solution of Eq. (10):

$$f_{\mathbf{1}} = \psi \left[ x_{\mathbf{1}} - \int_{0}^{z} \varphi(z) \, dz \right]. \tag{11}$$

The initial condition  $f_1(x_1; 0) = \psi_0(x_1)$  specifies a certain distribution for the random quantity  $X_1 = u_0^{"} - u_0^{'}$ . We will set the zero level z = 0 at the edge of the boundary layer,

where the core of the flow has been formed. Then  $f_1(x_1; 0)$  serves as a characteristic of the probable distribution of the random quantity  $X_1$  in the zone of developed turbulence in which the averaged velocity of the gas is independent of the transverse coordinate. Since even the highest crest of the wave on the film is still tightly bound to the main mass at the instant of separation, it might be assumed that the entire spectrum of possible values for  $u_0'' = u_0'$  is determined exclusively by the pulsations in the gas velocity  $u_0''$ . Results from the measurements of velocity pulsations [5] enable us to draw the conclusion that the appearance of any velocity out of some range of  $\Delta X_1 = x_{1max} - x_{1min}$  is equiprobable. The initial condition for Eq. (10) will then be

$$f_1(X_1; 0) = \begin{cases} \frac{1}{\Delta x_1} = \frac{1}{x_{1 \max} - x_{1 \min}}, & x_1 \in [x_{1 \min}, x_{1 \max}] \\ 0, & x \notin [x_{1 \min}, x_{1 \max}] \end{cases}.$$

The corresponding solution

$$f_1(x_1, z) = \begin{cases} \frac{1}{\Delta x_1}, & x_1 \in [x_{1\min}, x_{1\max}] \\ 0, & x_2 \notin [x_{1\min}, x_{1\max}] \end{cases}.$$
 (12)

Having substituted (12) into (9) and having carried out the replacement  $\alpha x_3 = \beta$ , we obtain the following expression for the density  $f_3(x_3)$ :

$$f_3(x_3) = \frac{1}{\Delta x_1} \int_{\frac{x_1 \min}{x_2}}^{\frac{x_1 \max}{x_2}} \alpha f_2(\alpha) d\alpha.$$
(13)

In this study we will not investigate the general properties of integral (13). Let us note only that this integral is equal to the constant in the interval  $[x_{3min}, x_{3max}]$  (outside of which it is equal to zero) independent of the distribution of the random quantities  $X_1, X_2$ ; this is valid for the functions  $f_2(x_2)$  such that integral (13) through direct integration reduces to a constant number. For example, for  $f_2(x_2) = const/x_2^2$ . Moreover, we can indicate a method of constructing a family of random quantities approximating a rather broad class of quantities  $X_2$ , so that  $f_3(x_3)$  is equal to the constant. Let us introduce the random quan-

tities y, assuming n values  $y_1 < y_2 < \ldots < y_n$  with probabilities  $p_1, p_2, \ldots, p_n(\sum_{i=1}^{n} p_i = 1)$  so that for k = 2, 3, ..., n,  $x_{1max}/y_k = x_{1min}/y_{k-1}$  and for k = 1, 2, ..., n,  $x_k p_k = \text{const.}$  It is obvious that for the random quantities y and for the limit random quantity (as  $n \rightarrow \infty$ )  $f_3(x_3)$  will be constant.

It is not difficult to note that, general speaking, integral (13) depends on the location of the ends of the distributions for the random quantities  $X_1$ ,  $X_2$ ,  $X_3$ . Specifically, if

$$\frac{x_{1\min}}{x_{3\min}} \leqslant x_{2\min} < x_{2\max} \leqslant \frac{x_{1\max}}{x_{3\max}},$$
(14)

then  $x_3$  is uniformly distributed. Let us estimate the values of the ends in our case:

$$\frac{x_{\rm imin}}{x_{\rm 3min}} = \frac{u_{\rm 0min} R}{v''} \left(\frac{R}{r'_{\rm 0max}}\right)^{-1} = \frac{u_{\rm 0min} r'_{\rm 0max}}{v''},$$

$$\frac{x_{\rm imax}}{x_{\rm 3max}} = \frac{u_{\rm 0max} R}{v''} \left(\frac{R}{r'_{\rm 0min}}\right)^{-1} = \frac{u_{\rm 0max} r'_{\rm 0min}}{v''}.$$
(15)

We will estimate  $r_{omin}$ ' and  $r_{omax}$ ' by using the results of observations into the behavior of the film. It has been demonstrated experimentally in [6] that in addition to large waves, a minor ripple also exists. It is natural to assume that these smallest drops are formed as a result of the separation of microwaves from the ripples, as in the atomization of a jet.



Fig. 1. Comparison of results from calculations according to formulas (16), (20), and (22) with the hydrograms from [9]. The distance from the film surface is 1.54-2.31 mm; ethyl alcohol: a)  $\bar{u}'' = 55 \text{ m/sec}$ ; b) 110.  $g(r_0')$ ,  $\mu m^{-1}$ ;  $r_0'$ ,  $\mu m$ .



Fig. 2. Comparison of the results for calculations according to formulas (16), (20), and (23) with the experimental data of [9] (the circles denote experiment): a) water; b) ethyl alcohol.  $r_0'$ ,  $\mu$ m; u", m/sec.

Following [7], we will write the condition for the wavelength  $\lambda$  of a low-viscosity liquid for which the wave will be unstable:

$$\lambda \geqslant \frac{2\pi\sigma}{\rho'' u_0^2}$$

The smallest possible unstable length is:

$$\lambda_{\min} = \frac{2\pi\sigma}{\rho'' u''^2}.$$

For the radius of the smallest drop we can therefore assume

$$r'_{0\min} = \frac{\lambda_{\min}}{4} = \frac{\pi\sigma}{2\rho'' \overline{u''^2}}$$
 (16)

If the small waves are formed as a result of the development of intrinsic oscillations in the film, the appearance of large overlapping waves is more reminiscent of analogous processes occurring on the surface of reservoirs under the action of the wind. For this case, [7] provides an analytical relationship between the wavelength  $\lambda$  and the local relative velocity u, which provides for its nonattenuating propagation:

$$\lambda = \frac{16\pi^2 \rho^{\mathrm{r}} v^{\mathrm{i}} u'}{c \, \rho^{\mathrm{r}} u^2} \, \lambda^{\mathrm{H}} \tag{17}$$

Here u' is the velocity of the liquid at the separation surface. Usually u' is regarded as the sum of the average flowrate and the mean phase velocity. Measurement results are presented in [8] for the average flowrate and phase velocity over a rather broad range of gas velocities u". With high u" these data can be generalized approximately by the relationship

$$u' \approx 0,083 \,\overline{u''}.\tag{18}$$

Substituting (18) into (17), in the case of c = 0.5 and  $u = \overline{u''} - u'$  we obtain

$$\lambda_{\max} = \frac{31,14\,\rho'\,\nu'}{\rho''\overline{\mu''}}.\tag{19}$$

For large waves, unlike the case of small waves, the relationship  $r_{0max}' = \lambda_{max}/4$  is considerably less accurate, since the amplitude of the larger waves at the instant of separation considerably exceeds the length of their bases. Such a definition of  $r_{0max}'$  enables us only to estimate the orders of magnitude. However, it is validated when the condition  $r_{0max}' \ge$  $r_{0min}'$  is observed, because in this case the change in the quantity  $r_{0max}'$  has little effect on the results of calculations carried out in accordance with formulas (17) and (18). Then, with consideration of (22), we have

$$r'_{0\max} = \frac{\lambda_{\max}}{4} = \frac{7,78\,\rho'\,\nu'}{\rho''\overline{u''}}.$$
(20)

Correspondingly, condition (14) can be rewritten in the form

$$\left(\frac{\pi}{2\rho''\sigma}\right)^{1/2} \frac{\rho' \,\mathbf{v}' \,u_{0\min}}{u_{0\max}} \leqslant \left(\frac{L_{\min}}{c_{\max}}\right)^{1/2} \leqslant \left(\frac{L_{\max}}{c_{\min}}\right)^{1/2} \leqslant \left(\frac{\pi^3 \,\sigma}{8\rho''}\right)^{1/2} u_{0\max}^{-1}.$$
(21)

Satisfaction of this condition ensures the constancy of integral (13) and, consequently, the uniform distribution of the random quantity  $X_3$ . If we assume that  $u_{0min}$  may be as small as desired, and  $L_{min}$  and  $L_{max}$  are on the orders of the corresponding radii, then with  $u_{0max}$  not overly large (16) will always be satisfied. Then, if  $X_3$  is distributed uniformly,  $X_3^{-1}$  is distributed in accordance with the law  $A/x^2$ . The radii of the forming drops are distributed in precisely the same way, and namely:

$$g\left(r_{0}'\right)=\frac{B}{r_{0}'^{2}}.$$

To determine the constants we can make use of the properties of distribution density

$$\int_{r_{0min}}^{r_{0max}} g(r_0') dr_0' = 1,$$

from which it follows that

$$B = \frac{r_{0\max} r_{0\min}}{r_{0\max} - r_{0\min}}$$

and

$$g(r'_{0}) = \frac{r'_{0\max} r'_{0\min}}{(r'_{0\max} - r'_{0\min}) r'_{0}^{a}}.$$
 (22)

For the mean radius we take the mathematical expectation

$$r'_{0} = \int_{1}^{r_{0}\max} g(r'_{0})r'_{0}dr'_{0} = \frac{r'_{0\max}r'_{0\min}}{r'_{0\max} - r'_{0\min}} \ln \frac{r'_{0\max}}{r'_{0\min}}.$$
 (23)

Calculations based on formulas (16), (20), (22), and (23), and their comparison with the experimental results of [9], are shown in Figs. 1 and 2. The increase in the divergence in the mean radii for water with the smaller  $\bar{u}''$  is associated with the violation of the condition  $r_{0max}' \gg r_{0min}'$  and, correspondingly, by the considerably greater influence of error in the determination of  $r_{0max}'$ .

## NOTATION

m, mass; *a*, acceleration;  $l(\tau)$ , length of the line bounding the plane of separation as a function of the time  $\tau$ ; L = l(0);  $\sigma$ , surface tension;  $F_{ad} = \sigma L$ , force of adhesion;  $\rho$ , density; u, velocity; v, kinematic viscosity; r', particle radius;  $r_{omax}$ ' and  $r_{omin}$ ', maximum and minimum possible particle radii; u", average gas flowrate; R, characteristic dimension. Superscripts ', ", and 0 pertain to the liquid, gas, and the instant of separation.

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## GAS PERMEABILITY OF NUCLEAR MEMBRANES AS A FUNCTION

OF EFFECTIVE PORE DIAMETER

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We have examined experimentally and theoretically the transport of various gases in channels with a diameter comparable to that of the molecule dimensions.

Interest in the processes of gas transport in membranes with ultrasmall pores exhibiting diameters of D < 10 nm is associated with the possibility of achieving an optimum combination of their permeability with high selective properties. Such membranes may result, for example, as an intermediate product in the production of quartz glass [1]. However, the indeterminacy of the geometric structure of porous glasses limits their selectivity and hinders interpretation of experiments [1] into the gas permeability at the microscopic level of the description.

In recent times so-called nuclear membranes (filters) have gained ever-increasing acceptance, and these filters are produced through the irradiation of polymer films with fission fragments in nuclear reactions [2] or by heavy ions in charged-particle accelerators [2-4], and the subsequent chemical etching of the tracks. Pores most nearly cylindrical in shape are obtained here with limited dispersion by size as a consequence of the above-described method by irradiation of films made of polyethylene terephthalate (PETP) [2]. The regular pore geometry of such a nuclear filter, given their ultrasmall dimensions, comparable to the characteristic dimensions of the potentials of intermolecular interactions, offers a unique possibility of establishing the relationship between the parameters of gas permeability through the filter and the microscopic characteristics of the gas and of the channel surfaces.

Isothermal (T  $\approx$  300 K) measurements of the permeability of He, Xe, Ar, and N<sub>2</sub> have been conducted in the present study on two specimens of nuclear PETP membranes whose pore diameters ranged in the interval 1.5-10 nm; these measurements have been carried out on these membranes in a series of "load-unload" cycles [5]. The experimental relationship of the relative permeability and the pore dimension is interpreted on the basis of a theoretical approach such as that covered in [6-8], within the scope of which we derive a solution to the system of interconnected kinetic equations for the functions of the distribution of free and adsorbed gas molecules within the channel.

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