The error due to instability of the light source is multiplicative and, therefore, $(u^+_{max} - u^+_{min})/u^+_{max}$ is independent of the light-source drift.

If solid spheres are obtained as a result of the monodisperse breakup of a jet, as when a liquid-metal jet breaks up and the resulting drops cool during flight, a weighing method based on the use of fiber-optic microbalance can be proposed.

The use of such a balance showed that when 500-μm-diameter lead spheres are weighed the relative error is 0.15. This can be used to reliably study the diameter distribution function of spheres obtained in modern drop generators.

Another parameter that affects the monodispersion of the induced breakup of a jet is the level of pressure pulsations in the drop generator. A fiber-optic pressure sensor [4], whose diagram is shown in Fig. 3, can be used to measure this level.

In summary, these fiber-optic sensors can be used to operationally monitor the jet parameters that affect the monodispersion of the jet breakup and to measure the diameter of the drops produced.

NOTATION

d_m, diameter of the manometer tube; d_{wg}, waveguide diameter; d_c, capillary diameter; u, sensor signal; u_{max}, maximum sensor dignal; u_{min}, minimum sensor signal; u_{dark}, sensor output signal that depends on the photodiode dark current; k, photodiode sensitivity; Φ, luminous flux; b, ratio of dark signals; T, photodiode temperature; and u^+, reduced sensor signal.

LITERATURE CITED

1. V. A. Grigor'ev, Vestn. Akad. Nauk SSSR, No. 4, 84 (1987).
2. Yu. D. Tretyakov, N. N. Oleinikov, and A. P. Mozhaev, Fundamentals of Cryochemical Technology [in Russian], Moscow (1987).
3. V. P. Ogorodnikov and V. V. Osipov, Sb. Nauch. Tr. Mosk. Energ. Inst., No. 149, 103-110 (1987).
4. V. G. Zhilin, Yu. P. Ivochkin, V. P. Ogorodnikov, and V. V. Osipov, Teplofiz. Vys. Temp., No. 5, 54 (1979).

CAPILLARY BREAKUP OF A LIQUID-METAL JET IN AN OXIDIZING MEDIUM

B. V. Artem'ev and S. G. Kochetov

UDC 532.5:66

The effect of oxidation of a free liquid-metal jet on its capillary stability is considered.

We have studied how oxygen impurity in a gas surrounding a thin jet affects the capillary stability of the jet. Our work was prompted by an experiment in which an attempt was made to obtain monodisperse capillary breakup of a gallium jet with diameter 0.5 mm, velocity 7 m × sec^{-1}, and temperature 60°C, flowing into argon that contained oxygen impurity. The jet did not break up at all if the oxygen concentration was higher than 0.2-0.3%. The transition from normal breakup to complete "non-breakup" occurred discontinuously as the oxygen concentration varied smoothly.

Below we report the experimental data and analysis of the effect that oxygen impurity in the surrounding atmosphere has on the capillary breakup of thin liquid-metal jets. We propose a model of this effect, based on the assumption that an oxide film is formed on the surface of the jet by the reaction of the jet with the oxygen in the surrounding medium. Since the oxide film presumably gives the surface elastic properties, a pressure counteracting the curvature of the surface arises along with the capillary pressure and can protract or completely stop capillary breakup.

Translated from Inzhenerno-fizicheskii Zhurnal, Vol. 60, No. 4, pp. 566-571, April, 1991. Original article submitted July 31, 1990.

0022-0841/91/6004-0425$12.50

Our experimental apparatus was built to obtain quantitative dependences of the length of the intact part of the jet on the percentage oxygen concentration in the surrounding medium, the discharge velocity, and the nozzle diameter. The description of the mechanism by which the oxygen impurity in the surrounding medium affects the capillary stability of a free liquid-metal jet combines three models describing: 1) the effect that the elastic oxide film on the jet surface has in increasing perturbations; 2) the formation of an oxide film; and 3) the formation of a global elasticity of the oxide film.

These models are considered successively and then combined for quantitative estimates of the effect of the oxygen in the surrounding medium on the capillary stability of the liquid-metal jet.

Effect of the Elastic Oxide Film on the Growth of Capillary Perturbations in the Jet. We consider a liquid-metal jet flowing into a motionless inert gas that contains oxygen impurity. The metal of the jet can be oxidized, the reaction is irreversible, and the reaction rate is rather high. The outflowing jet has a velocity u_0, which is constant along the length and the radius.

We consider the stability of a cylindrical liquid jet, with an oxide film on its surface, up to capillary breakup. For this purpose we use the Euler equation

$$\frac{\partial \mathbf{V}}{\partial \tau} + (\mathbf{V}\nabla)\mathbf{V} + \nabla P = 0, \quad \operatorname{div}\mathbf{V} = 0,$$

with boundary conditions

$$\frac{\partial u}{\partial y} = 0 \quad (y = y_0); \quad v = 0 \quad (y = 0);$$

$$P_c = \sigma\left(\frac{1}{y_0} - \frac{\partial^2 y_0}{\partial x^2}\right); \quad P_{el} = \frac{\overline{H} G(q)}{y_0^2}.$$

The pressure on the surface is determined, first, by the capillary pressure and, second, by the reaction of the oxide film on a random small perturbation $\overline{H}$ of the surface. Special "hanging-drop" experiments determined that the oxide film does not possess surface-active properties, i.e., does not cause the surface tension to decrease or to increase. Using the familiar technique for solving stability problems, we obtain the dispersion relation

$$C^2 = \frac{\sigma\left[(\alpha y_0)^2 - 1 + G(q)/\sigma\right] I_1(\alpha y_0)}{y_0 \rho \alpha I_0(\alpha y_0)}, \quad C = C_r + iC_i. \tag{1}$$

Jet breakup is impossible if

$$(\alpha y_0)^2 + G(q)/\sigma \geqslant 1. \tag{2}$$

Formation of an Oxide Film. The oxygen flow I(x) to the surface of the jet is determined from the description of the aerodynamics and mass transfer in a gas by equations of the "boundary layer" type. We used the integral equation method to obtain the solution. We assumed that the oxygen in the metal of the jet is on the saturation line and that the oxygen, diffusing toward the jet, goes into the bound state, forming an oxide film:

$$I(x) = 2D_g \rho_0 S^{-1}; \quad \frac{S^3}{45} + \frac{S^2}{10} = \frac{2x}{\mathrm{Re}}. \tag{3}$$

The surface density of oxygen on the jet is

$$q(x) = 16{,}9 N (\rho_0 u_0)^{-1} \int_0^x I(x)\, dx. \tag{4}$$

The most probable mechanism of surface oxidation [1] is one which produces not separate oxide molecules but island formations, whose total area increases with distance from the nozzle edge to the joint if this is not preceded by capillary breakup. On reaching the clean metal surface, the oxygen causes the area of the oxide film to increase. As the oxide-film fraction of the surface increases, however, the oxide film absorbs more oxygen which does not increase the area of the film but diffuses in beneath the film. Taking this into account, we write

$$M = 1 - \exp\left[-q(x) S_m\right]. \tag{5}$$

Elasticity of the Oxide Film. We consider a jet with a system of island oxide films. Individual islands or small clusters of them cannot have an appreciable elastic effect on the jet. As their area grows the islands join up into extensive conglomerates and form a global unified structure, which can react elastically to surface perturbation. For quantitative estimates we model the oxidizing surface of a jet of a continuous surface of pure metal, coated with equidistant round oxide islands of the same radius. No contact exists between the islands and the film does not have global elasticity when the radius of the islands is smaller than the determined value. As the oxidation proceeds the islands increase and link up. Starting from this time the surface elasticity of the film grows rapidly and reaches the elasticity of a continuous film. From the geometrical representation of the oxide film as a system of islands of the same radius we can easily estimate the relative area of the oxide film at which the oxide islands begin the link-up ($M_{in} = 0.85$) and the relative area at which the link-up is virtually complete ($M_c = 0.92$).

In our discussion below we introduce the average values of the surface density of oxygen on the jet that correspond to the beginning and end of the link-up. For this purpose we use M_c and M_{in} in (5) and take the size of the Ga_2O_3 molecule to be 4 Å, then $q_{in} = 0.38$ atom/Å^2 and $q_c = 0.43$ atom/Å^2.

On the basis of the above discussion, we approximate the elasticity of the oxide film with a step function

$$G(q) = \begin{cases} 0, & q < q_{in}; \\ G_0\left(\dfrac{q - q_{in}}{q_c - q_{in}}\right), & q_{in} \leqslant q < q_c; \\ G_0\left(\dfrac{q}{q_{in}}\right), & q \geqslant q_c. \end{cases}$$

The first relation in the approximation reflects the absence of surface elasticity of the oxide film when oxide films exist independently, the second reflects the rapid growth of elasticity as the islands link up, and the third reflects the linear growth of the surface elasticity of the continuous oxide film with increasing film thickness; G_0 is indeterminate and will be estimated below from the empirical fact that a stepped transition exists from the normal breakup of the jet to non-breakup with a smooth variation of the oxygen concentration of the surrounding medium.

A computational method for a quantitative analysis of the effect of oxygen impurity in the medium surrounding the jet has been developed on the basis of the models of oxide film formation, the initiation of surface elasticity, and the effect of an elastic oxide film on the capillary stability of the jet. The diameter and discharge velocity of the jet and the physical properties of the surrounding gas are the initial data and the elastic modulus G_0 of the oxide film is the parameter.

The condition for breakup is taken to be that the developing perturbation of the oxidizing jet must exceed the pre-breakup perturbation of the unoxidized jet at the maximum growing wavelength:

$$\alpha \int_0^x C_i dx \geqslant \alpha_{max} C_i L_0.$$

The initial perturbation is assumed to be independent of oxidation. Equation (2) is taken to be the condition for the end of the breakup.

The calculation yields the dependence of the length of the intact part of the jet on the oxygen concentration in the surrounding medium.

Figure 1 shows the characteristic calculated dependence of the length of the intact part of the jet flowing from a 0.5-mm-diameter hole with velocity 7 m/sec at $G_0 = (10, 1.0, 0.1)\sigma$. We see that the shape of the curves reflects the empirical finding that there is an abrupt transition from normal breakup to non-breakup as the oxygen concentration in the carrier gas varies smoothly only at $G_0 \geq \sigma$. This will be the lower estimate of the elastic modulus of the oxide film, which is sufficient for carrying out calculations from the proposed model.

Experimental Apparatus. The experimental apparatus is intended for studying the quantitative effect of oxygen impurity on the capillary instability. Its main components are (Fig. 2): a feeder tank 1 fitted with an electric heater to store, heat, and pressure-transfer Ga into the jet-

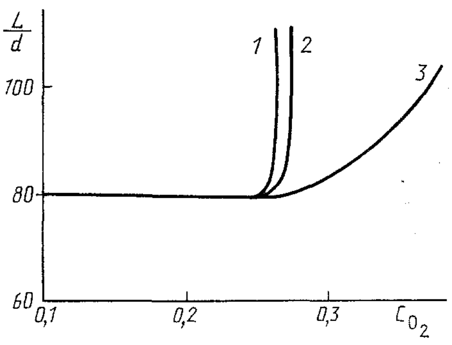

Fig. 1

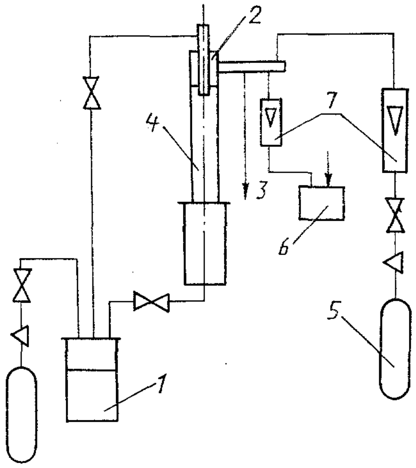

Fig. 2

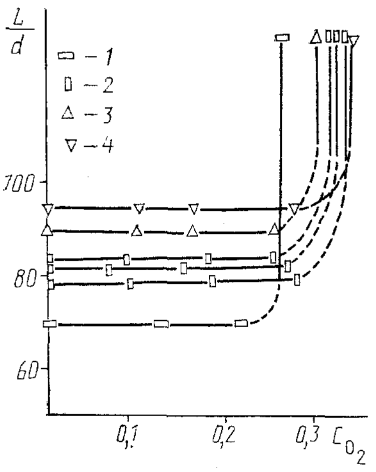

Fig. 3

Fig. 1. Graph of calculated length of the intact part of a jet with diameter 0.5 mm and velocity 7 m/sec [L/d is the relative length of the intact part of the jet; C_{O_2} is the oxygen concentration in the surrounding gas (argon), %]; 1) $G_0 = 10\sigma$; 2) 1.0σ; 3) 0.1σ.

Fig. 2. Experimental apparatus.

Fig. 3. Experimental dependence of the length of intact part of jet on oxygen concentration in the surrounding gas at various jet velocities: 1) 3.2; 2) 4.95; 3) 5.5; 4) 9.2 m/sec.

forming unit; a discharge unit, 2, equipped with a piezoelectric crystal to impose the prescribed perturbations in the jet, for forming the jet and measuring the gallium temperature immediately before outflow as well as taking gas samples for the gas analyzer 3; a glass tube 4 for forming a gas atmosphere around the jet; and a gas system for preparing the gas atmosphere and feeding it to the forming unit. The gas system has a bottle 5 of argon, a compressor 6 for feeding atmospheric air into the gas line, rotameters 7 for measuring the flow rate of the carrier gas and the atmospheric air, and a line for reliable mixing.

The gallium temperature was measured and monitored with chromel-alumel thermocouples, installed hermetically in the discharge tank (3) and in the discharge unit (1). The length of the intact part of the jet was measured with an N-360 cathetometer and was recorded with a ZENIT-E camera. The oxygen concentration in the carrier gas was measured with a GAZOKhROM-3101 gas analyzer. A gas sample was taken for this purpose with a medical hypodermic syringe in the direct proximity of the discharge unit. The jet velocity was determined from the distance between drops and the frequency of the perturbations.

Draw dies with holes with diameters of 0.3, 0.5, and 0.7 mm and length (die thickness) of 0.2 mm were used in the experiment.

Experimental Results and Conclusion. Figure 3 shows the results of an experiment with jets of diameter 0.5 mm. The curves obviously change abruptly in the region of a 0.2-0.3% oxygen concentration in the surrounding medium. This is in good agreement with calculations with the proposed method.

The region of the oxygen concentration below the dashed part of the curve corresponds to a pulsating breakup. With increasing oxygen concentration in the surrounding medium from zero to a value C_1 breakup occurs without changes. With the concentration C_1 the jet breakup begins to pulsate, normal breakup is replaced irregularly by nonbreakup and conversely at a frequency of about 1 Hz. As the oxygen concentration rises the fraction of time of normal breakup decreases and at the concentration C_2 the jet does not break up at all. The pulsating mode of breakup corresponds to a rather narrow range of concentrations of ~0.05% and is attributable to the random nature of the formation of the oxide film from separate islands.

Our criterion for generalizing the experimental data was the surface concentration of oxygen on the jet at which the pulsating mode ends and no breakup at all occurs. The oxygen flow to the jet was determined from (1), using the experimentally determined length of the intact part of the jet as a function of the oxygen concentration in the carrier gas, and the oxygen concentration on the jet is found from (4).

TABLE 1. Experimental Results

Jet diam, mm	Discharge velocity, m/sec	Oxygen concn. in the gas,* %	q_{in}, atom/Å^2	q_c, atom/Å^2
0,3	7,7	0,21—0,24	0,34	0,41
0,3	9,1	0,26—0,31	0,32	0,39
0,3	13,9	0,29—0,33	0,32	0,40
0,5	3,2	0,21—0,26	0,33	0,39
0,5	4,9	0,25—0,32	0,33	0,40
0,5	5,0	0,26—0,32	0,34	0,41
0,5	5,0	0,23—0,30	0,34	0,41
0,5	5,5	0,25—0,30	0,34	0,41
0,5	9,2	0,27—0,33	0,33	0,39
0,7	2,9	0,20—0,25	0,31	0,38
0,7	4,5	0,21—0,24	0,34	0,42
0,7	5,8	0,22—0,27	0,34	0,42
0,7	6,0	0,22—0,26	0,32	0,40
0,7	6,0	0,23—0,26	0,33	0,41

---

*Indicated concentrations of the initiation of the pulsating mode with no breakup.

Table 1 shows the results of our experiment with jets with diameter 0.3, 0.5, and 0.7 mm under induced breakup. We see that over a wide range of parameters (jet diameter, discharge velocity, with an appreciable difference in the length of the intact part) the calculated oxygen concentrations on the jet are similar for all the modes and amount to $q_{in} = 0.31\text{-}0.34$ atom/Å^2 and $q_c = 0.38\text{-}0.42$ atom/Å^2.

The proposed model can be refined by using the experimental value of the surface density of oxygen on the jet for which no breakup occurs. As the indicated density we took the oxygen density that results in no breakup at all, 0.403 atom/Å^2.

The dependence of the intact part of the jet on the concentration of oxygen impurity in the surrounding gas was calculated with the proposed model, using the empirically refined values of the oxygen density on the surface of the jet for which no breakup occurs; the results obtained in this way are close to the experimental data.

NOTATION

x and y, longitudinal and radial coordinates; y_0, radius of the jet; P_c, capillary pressure; P_{el}, elastic reaction of the oxide film; $2\pi\alpha^{-1}$, perturbation wavelength; σ, surface tension of the jet material; S, thickness of concentration losses; G, elastic modulus of the oxide film; q, surface concentration of oxygen on the jet, atom/Å^2; D_g, gas diffusion coefficient; ρ_0, oxygen density under normal conditions; ρ, density of the jet material; S_m, area of one oxide molecule; N, Avogadro number; and M, fraction of the jet surface occupied by oxide film.

LITERATURE CITED

1. V. P. Zhdanov, Elementary Physicochemical Processes on a Surface [in Russian], Novosibirsk (1988).