

UNSTEADY MOTION OF DROPLETS IN A SYSTEM OF TWO IMMISCIBLE LIQUIDS

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The accelerated motion of droplets after their formation, the motion of droplets in the pre-coalescence period and the motion of oscillating droplets moving through an motionless continuous phase have been described using a simplified concept of the droplet as a mass point and a concept of the motion as that of an ellipsoidal droplet in an ideal fluid. The values calculated from the derived equations have been compared with experimental data obtained from a motion-picture camera record of the motion of the droplets in several two-phase, equilibrium, low-viscosity and high-grade purity Newtonian systems. In most cases water was the dispersed phase. The experiments included tests of the so far published descriptions of the behaviour of oscillating droplets.

The research of the processes in liquid dispersions of technical importance necessitates the knowledge of the laws governing the behaviour of individual droplets. It turns out that the behaviour of droplets even under relatively simple situations is complex and has not been fully comprehended and described. The lack of information is particularly severe as far as the hydrodynamic part of the motion of the droplet is concerned which is important for the description of mass or energy transfer across the droplet's interface. Among the most important characteristics belong velocity and the nature of the shape changes of the droplet. The gap appears most critical in the description of the unsteady motion of the droplets which takes place in the initial and the final stage of the droplet's life and mainly as the so-called oscillations occur. The main aim of this work has been to examine the transients of the motion of the droplets in these situations.

The droplet after its formation moves in an accelerated manner until the forces of the hydrodynamic resistance against the motion and the gravity force are balanced. This period of the droplet's life shall be called the stage of accelerated motion. If the droplet during its motion changes periodically and its shape remains axially symmetric, its velocity fluctuates about a certain mean designated as the velocity of the quasi-steady motion, v_t . On hitting the interface, which can be either deformable or rigid, the droplet experiences for some time damped unsteady motion until it disappears by coalescence.

This paper is a continuation of the preceding communication^{1,2} where it was shown that the shape changes of oscillating droplets are surprisingly highly reproducible and that in the stage of

quasi-steady motion the shape changes are markedly more intensive than those in the stage of accelerated motion. The velocity of the quasi-steady motion, v_t , and the period of oscillation, τ , in our systems can be estimated from existing^{3,4} empirical correlations (see Eqs (1) and (2)). These correlations were formulated from experiments with other systems where the continuous phase was always water.

$$v_t = D[(0.535 - 0.324 \Delta \varrho / \varrho_D) \tau_L]^{-1}, \quad (1)$$

where

$$\tau_L = \frac{\pi}{4} \left[\frac{3\varrho_D + 2\varrho_C}{3\sigma} D^3 \right]^{1/2}$$

and

$$\tau = \left\{ \frac{1}{2\pi} \left[\frac{192\sigma}{(3\varrho_D + 2\varrho_C) D^3} \right]^{1/2} - \frac{0.695}{D^2 (\Delta \varrho / \varrho_D)^{0.2}} \right\}^{-1}. \quad (2)$$

This work concerns the transients of the motion of the droplets in the stage of accelerated motion and the stage of quasi-steady motion examined separately. The theoretical description is formulated using highly simplified concepts.

THEORETICAL

The motion of the droplets in the stage of accelerated motion. It is assumed that a) the droplet moves as a mass point and the resistance within the continuous phase is proportional to the instantaneous velocity of the droplet. b) In the steady state the droplet moves at the velocity v_t given by Eq. (1).

The equation of motion of the mass point takes the form

$$\varrho_D V_F \ddot{y} = \Delta \varrho g V_F - k \dot{y}. \quad (3)$$

The initial conditions are

$$v(t=0) = \dot{y}(0) = 0, \quad \lim_{t \rightarrow \infty} v(t) = v_t. \quad (4), (5)$$

The solution is sought in the form

$$v(t) = v_t \left[1 - \exp \left(- \frac{\Delta \varrho g t}{\varrho_D v_t} \right) \right]. \quad (6)$$

The quasi-steady motion of oscillating droplets. It is assumed that a) the flow within and without the droplet steadies down much faster in comparison with the shape changes of the droplet. b) The resistance against the motion of the droplet is directly proportional to the velocity of its motion and the induced mass. c) The losses of kinetic energy by friction in both phases are negligible. The induced mass

can be characterized by the tensor of induced mass, M_{ij} , related (see *e.g.* ref.⁵) to the kinetic energy, K , of the liquid forming the continuous phase by

$$K = \frac{1}{2} v_i v_j M_{ij}, \quad (7)$$

v_i is a component of the velocity vector of the motion. If it is assumed for simplicity that the shape of the droplet in each time instant may be approximated by rotational ellipsoid of equal volume and the ratio E of the height to the width (the axis of symmetry of the ellipsoid is parallel to the direction of motion of the droplet), the only non-zero principal component of the tensor of induced mass, M_y , may be written as^{5,6}

$$K = \frac{1}{2} \dot{y}^2 M_y, \quad (8)$$

where

$$M_y \equiv \rho_c V_F \mathcal{F}(E). \quad (9)$$

The kinetic energy, K , of the motion of the rotational ellipsoid of ideal fluid can be computed from

$$K = -\frac{\rho_c}{2} \iint_{\mathcal{S}} \Phi \frac{\partial \Phi}{\partial n} d\mathcal{S}, \quad (10)$$

where it is integrated over the surface of the ellipsoid \mathcal{S} ; Φ is the corresponding velocity potential and n is the normal to the surface \mathcal{S} . On integrating (the integration is dealt with in detail in the monograph listed as ref.⁷ and on combining Eqs (8) and (9) the dependence of the function \mathcal{F} on the shape characteristic E is obtained in the form

$$\mathcal{F}(E) = \begin{cases} \frac{1}{E} \frac{\sqrt{(1-E^2)} - E \arcsin \sqrt{(1-E^2)}}{\arcsin \sqrt{(1-E^2)} - E \sqrt{(1-E^2)}}, & E < 1 \\ \frac{1}{E} \frac{\sqrt{(E^2-1)} - E \operatorname{arccosh} E}{\operatorname{arccosh} E - E \sqrt{(E^2-1)}}, & E > 1 \end{cases} \quad (11)$$

From the assumptions *a)–c)* it follows that the instantaneous velocity, $v(t)$, is related to its instantaneous shape, $E(t)$, and the average shape, \bar{E} , by

$$v(t) \cdot \mathcal{F}[E(t)] = \bar{v} \mathcal{F}(\bar{E}), \quad (12)$$

where

$$\bar{v} = \frac{1}{\tau} \int_{t_0}^{t_0+\tau} v(t) dt, \quad \bar{E} = \frac{1}{\tau} \int_{t_0}^{t_0+\tau} E(t) dt. \quad (13)$$

With the knowledge of the constant \bar{v} and \bar{E} and the course of the function $E(t)$ one can compute the course of the function $v(t)$. The resulting relation (12) describes the motion of an ellipsoidal droplet in an ideal fluid. Similarly we could proceed in case of other shapes assumed for the droplet than the rotational ellipsoid; the function \mathcal{F} then, of course, takes a different form.

EXPERIMENTAL

Individual droplets, formed reproducibly, moved through a vertical cylindrical glass column through the wall of which were filmed by a high-speed motion picture camera. The evaluation of the obtained pictures yielded time dependences of the velocity of motion of the droplets. The apparatus and the experimental technique have been described in more detail elsewhere^{1,2,8}. The experiments were carried out with the following systems. The first named phase was dispersed: water-n-hexane; water-cyclohexane; water-n-heptane; water-benzene; water-diethyl ether; water-aniline; tetrachloromethane-water; aniline-n-heptane. The used substances were of *p.a.* grade and were purified before use by repeated rectification. All experiments were carried out at 25°C and the apparatus as well as the solvents were carefully kept clean during experiments.

RESULTS AND DISCUSSION

The measured velocities of the droplets in the stage of accelerated motion were compared with the values computed from Eq. (6). The comparison for the water-n-hexane systems is shown in Fig. 1. The curve I corresponds to the theoretical curve, Eq. (6), and the broken line II indicates the velocity of the quasi-steady motion, v_v ,

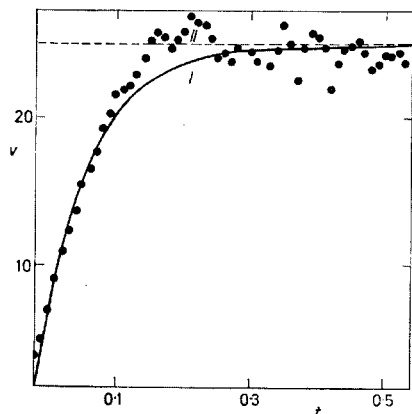


FIG. 1
Transient Development of Velocity of Droplets in the Water-n-Hexane System in the Stage of Accelerated Motion

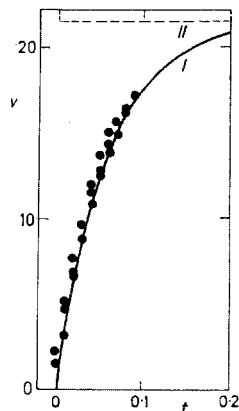


FIG. 2
Transient Development of Velocity of Droplets in the Tetrachloromethane-Water System in the Stage of Accelerated Motion

from Eq. (1). Individual points indicate the velocity of the leading face of the droplet averaged over the time interval of 0.04 s. The same situation for the tetrachloromethane–water system is shown in Fig. 2 and for the aniline–*n*-heptane system in Fig. 3. Similar results were obtained also with the other systems. In the water–benzene system, where within certain limits the volume of the droplet almost does not depend on the rate of its formation, the droplet was generated at two different rates. Empty circles in Fig. 4 represent results with rapidly formed droplets (the time of generation of a droplet of volume 0.122 cm³ by discharging liquid from a nozzle 0.153 cm in diameter was 2 s), full circles concern the experiments with slowly generated droplets (time of generation about 30 s). From Fig. 4 it is apparent that the rate of generation of the droplet does not exercise any appreciable effect on the time dependence of the rate of its motion.

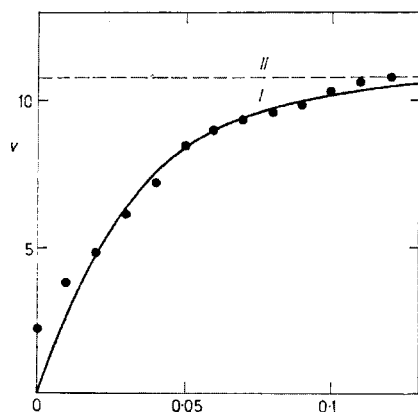


FIG. 3
Transient Development of Velocity of Droplets in the Aniline–*n*-Heptane System in the Stage of Accelerated Motion

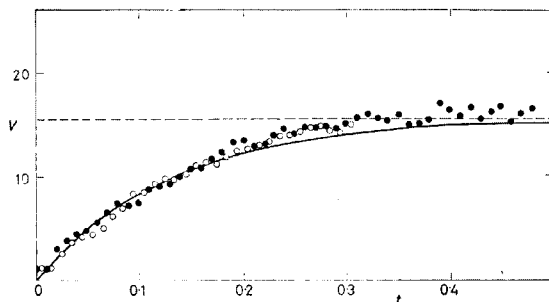


FIG. 4
Transient Development of Velocity of Droplets in the Water–Benzene System in the Stage of Accelerated Motion

○ Slowly and ● rapidly generated droplets.

The presented figures indicate that the calculated and measured time dependences of the velocity of motion of droplets well agree; systematic deviations appear toward the end of the period of acceleration. This is probably caused by the change of the character of the flow within and without the droplet which begins to intensively oscillate.

A sequence of shapes of an oscillating droplet during a single oscillation period is shown in Fig. 5 plotting also the time dependence of the velocity of the leading

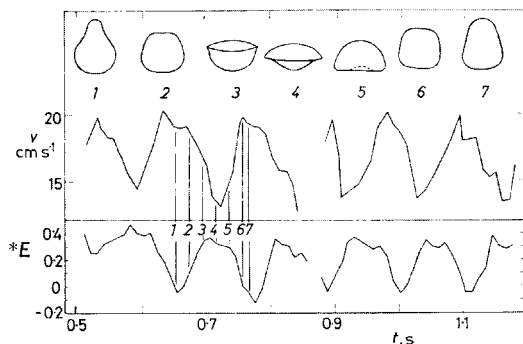


FIG. 5

Transient Development of Velocity and Shape Characteristic E^* of Droplets in the Water-Benzene System in the Stage of Quasi-Steady Motion

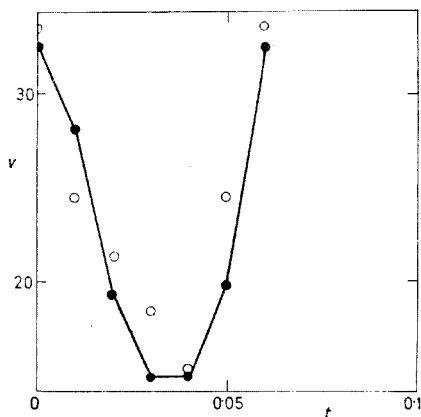


FIG. 7

Comparison of Computed and Measured Instantaneous Velocities of Droplets in the Water-n-Hexane System in the Stage of Oscillation

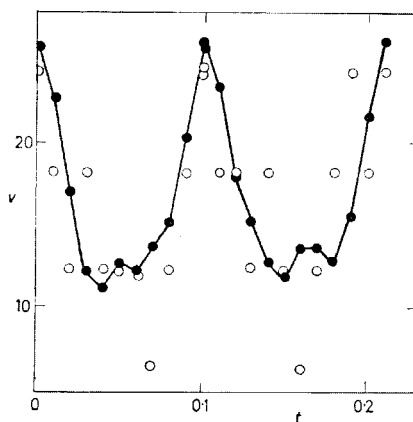


FIG. 8

Comparison of Computed and Measured Velocities of Droplet in the Water-Benzene System during Two Different Oscillation Periods

face of the droplet and the geometrical characteristics of its shape, E^* . The relation between individual phases of the shape change and the given time dependences has been expressed in a numerical form: The outlines of the droplets shown in Fig. 5 were traced from the film record of the experiment with a water droplet (volume 0.122 cm^3) moving in benzene. Part of the film record is shown for illustration in Fig. 6.* On this figure the time elapsed between individual frames is 0.01 s . The shapes of oscillating droplets in the other systems were very similar. A comparison of the measured transients of the velocity of motion of the droplets with those computed from Eqs (11) and (12) is furnished in Fig. 7 for the water–n-hexane systems (empty circles designate measured values, full circles the calculated values). The same dependences for two oscillation periods of the droplet in the water–benzene systems are shown in Fig. 8. Here the real velocity at the time of local minimum is significantly lower than the corresponding computed value. At this instant the shape of the droplet (see shape No 5 in Fig. 5) is markedly different from that of the ellipsoid. During the motion of the droplet the concave part of its surface induces the flows of the continuous phase oriented against the motion of the bulk of the ambient liquid with correspondingly large resistance against the motion of the droplet and lower velocity. Quantitatively, this effects could be estimated in essentially the same manner, except, of course, using a more realistic approximation of the shape of the droplet. In all examined systems the values defined by Eq. (13a) and v_1 from Eq. (1) were very close. It turned out that as long as we substitute v_1 for \bar{v} in Eq. (12) we obtain practically identical results.

In the same manner as that used in the stage of accelerated motion we have also examined the motion of the droplets in the pre-coalescence stage. We found out that the vertical damped oscillations occurring after hitting the interface too can be to some extent described with the aid of the model viewing the droplet as a mass point (see Appendix).

The frequencies and the damping of the amplitudes of the oscillations of the droplets in the stage of accelerated motion and in the pre-coalescence period were compared with the values following from the theoretical or empirical relations published in the literature. The observed time periods of the shape changes in the pre-coalescence period are shown for droplets of different size and the water–benzene system in Fig. 9. The curve I in this figure represents theoretical prediction of Miller and Scriven⁹ and the curve II an empirical correlation derived for the free motion of the oscillating droplets by Schroeder and Kintner¹⁰. A similar comparison for the same system and the oscillations in the stages of accelerated and quasisteady motion is shown in Fig. 10. Unlike the stage of quasi-steady motion, when the frequency of oscillations is well described by the correlation proposed by Edge and Grant⁴ (Eq. (2)), the

* See insert facing page 3536.

stage of accelerated motion (similarly as in the pre-coalescence period) is best described by the expression of Miller and Scriven⁹. The same conclusion was obtained also for the other systems (see the comparison of computed (τ_C) and measured (τ_E) periods) in the stage of accelerated motion furnished in Fig. 11. This is interesting

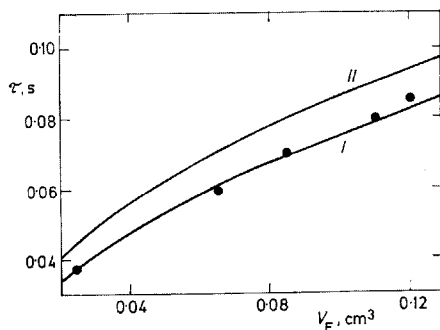


FIG. 9

Comparison of Computed and Measured Time Periods of Oscillations in the Water-Benzene System in the Pre-Coalescence Stage

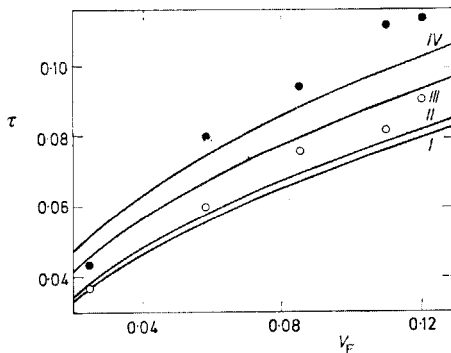


FIG. 10

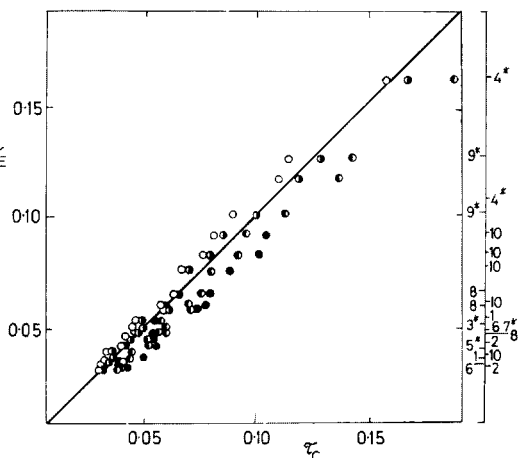
Comparison of Computed and Measured Time Periods of Oscillation in the Water-Benzene System in the Stage of Accelerated Motion (\circ) and the Stage of Quasi-Steady Motion (\bullet)

The lines represent relations published by Lamb¹¹ (I), Shroeder and Kintner¹² (III), Miller and Scriven⁹ (II), and Edge and Grant⁴ (IV).

FIG. 11

Comparison of Measured Periods of Shape Changes in the Stage of Accelerated Motion with the Values Following from Equations

Published by Lamb¹¹ (\circ), Miller and Scriven⁹ (\bullet), Shroeder and Kintner¹² (\odot), and Edge and Grant⁴ (\bullet). The numbers on the right hand side of the figure indicate the system used: 1 water-n-hexane, 2 tetrachloromethane-water, 3 water-n-butanol, 4 aniline-water, 5 aniline-n-heptane, 6 water-n-heptane, 7 water-diethyl ether, 8 water-cyclohexane, 9 water-cyclohexanol, 10 water-benzene. The asterisks indicate the case when the empirical correlation of Edge and Grant⁴ yielded a value differing by more than several tens of percent.



due to the fact that in the stage of accelerated motion as well as in the pre-coalescence period the amplitude of oscillations is considerably greater than admitted by the assumptions of Miller's and Scriven's theory (the amplitudes of oscillations in the pre-coalescence stage are practically just as large as those in the stage of quasi-steady motion as it is shown in Fig. 6). This suggests that decreased frequency of oscillations of the quasi-steady motion is rather the result of flow forces than the magnitude of the amplitude of oscillations. Similarly we also compared the characteristics of the damping of the oscillations in all stages of life of the droplet. These data too are well described by the expressions derived by Miller and Scriven⁹. A detailed description of these results is given elsewhere⁸.

APPENDIX

The following equation was used to describe the vertical motion of droplets after their impact on the liquid–solid horizontal wall interface

$$\rho_D V_F \ddot{y} = -\Delta \rho g V_F - k \dot{y}, \quad (14)$$

where

$$k = \Delta \rho g V_F / v_t,$$

with the conditions

$$y(t=0) = 0, \quad \dot{y}(t=0) = \alpha v_t. \quad (15)$$

Solution of this equation depends on the value of the parameter α , which was sought experimentally. In the water–benzene system its value amounts approximately to 0.8. The time course of the position of the droplet with respect to the level of the interface following from Eqs (14) and (15) agreed to within 10–25% with the observed values.

The motion of the droplet after impinging the liquid–liquid interface was described by the following equation

$$\rho_D V_F \ddot{y} = - \left(\frac{q}{t} \right) y, \quad (16)$$

with the conditions

$$y(t=0) = 0, \quad \dot{y}(t=0) = v_t. \quad (17)$$

For the solution, assumed in the form of an infinite power series (for derivation see *e.g.* ref.¹⁰), it follows from the above equations the transient position of the droplet. This course agreed in the water–benzene systems with the experimentally found values to within 5–20%.

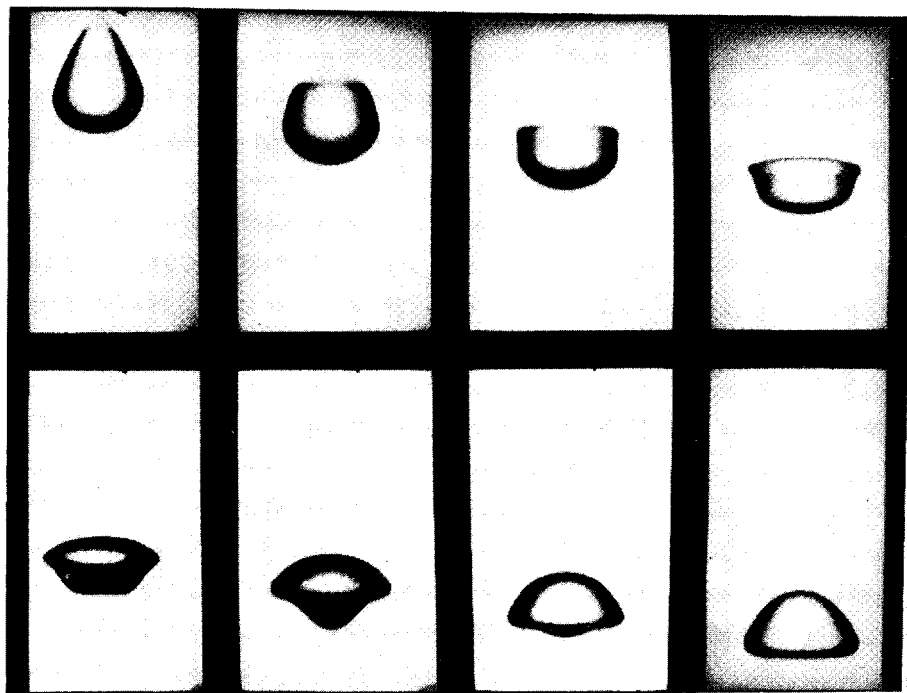


FIG. 6

Motion Picture Camera Photographs of Oscillating Droplet in the Water-Benzene System
Droplet is shown 8–10 cm below the mouth of droplet generating nozzle.

LIST OF SYMBOLS

E	characteristic of shape; ratio of maximum vertical and horizontal size of droplet or ellipsoid, $E^* = (1 - E)/(1 + E)$
\mathcal{F}	function characterizing form drag, defined in the text
g	acceleration due to gravity
$D = (6V_F/\pi)^{1/3}$	diameter of spherical droplet
k	constant
K	kinetic energy of continuous phase
M_{ij}	tensor of induced mass, defined in text
M_y	principal component of tensor of induced mass in direction of y
n	normal to element of surface
q	constant
\mathcal{S}	surface of body surrounded by flow of liquid
t	time [s]
v	velocity [cm/s]
v_i	component of velocity field in continuous phase
v_t	steady mean velocity of motion of oscillating droplet
V_F	volume of droplet
y	cartesian coordinate in direction of gravity force
$\dot{y} = dy/dt$	
ρ_c	density of continuous phase
ρ_D	density of dispersed phase
$\Delta\rho$	density difference of both phases
σ	interfacial tension
τ	period of oscillation
τ_L	period of oscillation according to Lamb ¹¹
ϕ	velocity potential
\varkappa	constant

REFERENCES

1. Vacek V., Nekovář P.: Presented at the IV-th International CHISA Congress, Prague, October 1972.
2. Dinh S. V., Nekovář P., Vacek V.: Presented at the IV-th International CHISA Congress, Prague, October 1972.
3. Thorsen G., Stordalen R. M., Terjesen S. G.: Chem. Eng. Sci. 23, 413 (1968).
4. Edge R. M., Grant C. D.: Chem. Eng. Sci. 26, 1001 (1971).
5. Landau L. D., Lifšic E. M.: *Mekhanika Sploshnykh Sred.* Moscow 1954.
6. Luiz A. M., Chem. Eng. Sci. 22, 1083 (1967); 24, 119 (1969).
7. Milne-Thomson L. M.: *Theoretical Hydrodynamics*, p. 500. MacMillan, London 1968.
8. Vacek V.: *Thesis*. Institute of Chemical Technology, Prague 1973.
9. Miller C. A., Scriven L. E.: J. Fluid Mech. 32, 417 (1968).
10. Jayaratne O. W., Mason B. J.: Proc. Roy. Soc., Ser. A 280, 545 (1964).
11. Lamb H.: *Hydrodynamics*, p. 473. Dover, New York 1932.
12. Schroeder R. R., Kintner R. C.: A. I. Ch. E. J. 11, 5 (1965).

Translated by V. Staněk.