Drop formation in a transient régime of dispersion

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THE OBSERVATIONS of liquid efflux through the circular orifices showed that there are two different régimes of drop formation for the relatively low flow rates: the first one when the drop is formed close to the orifice, and the second one when there exists a jet and the drop forms on its end. The aim of the present work has been the study of the transient régime between the two mentioned above. In the experiments performed, two immiscible liquids were used with almost the same density but very different viscosities — the liquid flowing out of the orifice had a much greater viscosity. The choice of these conditions was determined by the fact that the transient régime is in this case very wide, i.e. it occurs at different flow rates. The results of the experiments and simple mathematical models allowing for a physical interpretation are presented in this paper.

Obserwacje cieczy wypływających z kołowych otworów wykazały, że istnieją dwa różne obszary tworzenia się kropli przy stosunkowo niskich prędkościach przepływu: pierwszy, gdy kropla tworzy się tuż przy otworze, a drugi, gdy istnieje struga i kropla tworzy się na jej końcu. Celem niniejszej pracy było zbadanie obszaru przejściowego, występującego między dwoma wyżej wspomnianymi obszarami. W pizeprowadzonych doświadczeniach użyliśmy dwóch niemieszających sie płynów o prawie takiej samej gęstości, lecz różnych lepkościach (ciecz wypływająca z otworu posiadała dużo większą lepkość). Wybór tych warunków wynikał z faktu, że obszar przejściowy jest w tym przypadku bardzo szeroki, tzn. zachodzi dla różnych prędkości przepływu. W piacy tej przedstawione zostały wyniki doświadczalne i proste modele matematyczne, umożliwiające fizyczną interpretację zjawiska.

Наблюдения жидкостей вытекающих через круговое отверстие показали, что существуют две разные области образования капли при сравнительно низких скоростях течения: первая, когда капля образуется близко отверстия, и вторая, когда вытекает струя и капля образуется на ей конце. Целью настоящей работы являлось исследование переходной области, выступающей между двумя вышеупомянутыми областями. В проведенных экспериментах использованы две несмещивающиеся жидкости с почти такой самой плотностью, но с разными вязкостями (жидкость истекаемая из отверстия имеет много большую вязкость). Подбор этих условий следует из факта, что переходная область в этом случае очень широка, т. зн. она существует для разных скоростей течения. В этой работе представлены экспериментальные результаты и простые математические модели дающие возможность физической интерпретации явления.

Notation

- Bo = $\Delta gg D^2 / \sigma$ Bond number,
- D internal diameter of the orifice,
- d jet diameter,
- F force,
- $g = 981 \text{ cm/s}^2$ gravitational acceleration,
- h drop height at time of formation,
- $\overline{h} = h/D$ dimensionless height of a drop,
- K interface curvature,
- m drop mass,
- $\overline{m} = m/\varrho_d \pi D^3$ dimensionless mass of a drop,
- R radius of a drop treated as a sphere,

- $\overline{R} = R/D$ dimensionless radius of a drop,
- Re = $\varrho_d UD/\mu_d$ Reynolds number,
 - t time,
 - $\overline{t} = t U/D$ dimensionless time,
- U mean velocity of the efflux through the orifice,
- u velocity in a jet (mean value in the cross-section),
- V total volume of drop and jet (if the latter exists) at the time of formation,
- $\vec{V} = V/D^3$ dimensionless volume of drop and jet,
- v velocity of drop mass center,

$$\overline{v} = \frac{v}{U}$$
 dimensionless velocity of drop mass center,

We = $\varrho_d U^2 D / \sigma$ Weber number,

- x coordinate of mass center of (spherical) drop,
- $\overline{x} = x/D$ dimensionless coordinate of drop mass center,
- $\Delta \varrho = \varrho_d \varrho_c$ difference between densities of liquid phases,
 - θ angle between the tangent to drop surface and the direction parallel to drop axis of symmetry either in the cross-section connecting the drop and a jet or in the plane of the orifice (cf. Fig. 4b),
- μ liquid viscosity,

9)

- $\overline{\mu} = \mu_d/\mu_c$ ratio of viscosities,
- ę liquid density,
- $\overline{\varrho} = \Delta \varrho / \varrho_d$ dimensionless parameter,
- σ interfacial tension.

Lower indexes

- c, d refer to the continuous and dispersed phase, respectively,
 - 0 refer to the beginning of the 1st stage of the period of drop formation in the presented theory,
- 1, 2, 3 refer to the end of the 1, 2, 3rd stages of drop formation, respectively.

1. Introduction

THE FORMATION of drops in conditions when one liquid flows into another immiscible liquid through a circular orifice depends on many factors and among others on:

the way of flow extortion,

the material properties of the liquids and properties of the nozzle material,

the mean flow rate of a dispersed phase,

temperature conditions.

Most of the recorded experimental work on dispersion from circular orifices has been done in conditions of continuous injection of a dispersed phase and under constant temperature of the system [1-8]. IZARD *et al* [9] showed that the discontinuous injection could give (in certain conditions) a dispersion of very uniform drops. Correlation for the drop sizes resulting from a dispersion was developed when the continuous phase was static

[1, 2, 4, 5, 7, 8] and when it flowed in a counter current [3, 6]. Many findings on drop formation in liquid systems were applied in processes of extraction because of the significant influence of these phenomena upon the characteristics of equipment in the chemical industry [3, 10–15].

Drop formation and the breakup of a jet in liquid-liquid systems has also been treated theoretically [e.g. 5, 8, 16–19] but it seems that these efforts are still in the initial stage.

The purpose of some of our experiments [20, 21] was to examine the nature of drop formation within the range of the relatively low flow rates of a dispersed phase. The injection of this phase was continuous and the outer liquid (i.e. continuous phase) was static. The three liquid systems studied were chosen in such a way that two liquids of every pair had almost the same density but very different viscosities (detailed experimental data are given in [21]). Such a choice of liquid systems gives an increase of the geometrical scale of observed effects e.g. dimensions of drops and jets. Another advantage is that the experimental procedure is greatly simplified. These experiments confirmed some qualitative observations described in the literature [2, 5, 6, 22, 23] concerned with the changes in the mechanism of drop formation occuring under variation of the dispersed phase flow rate.

It is well-known from observations performed at low flow rates that there are two main different régimes of drop formation: the first one occurs when the drop is formed close to the orifice, and the second one is when there exists a jet and the drop forms at its end. The places of these régimes can be pointed at in a convenient way on the diagram (taken

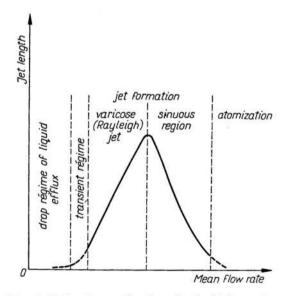


FIG. 1. Jet length as a function of a liquid flow rate.

from [23]) where the jet length is the only function of a mean flow rate through the orifice (Fig. 1). In the previous work [21] particular attention was paid to the transient régime between the two mentioned above. It was noted that this régime is especially wide (i.e. it

occurred for different flow rates) in the systems where the liquid flowing out of the orifice had a much greater viscosity than the outer liquid (continuous phase).

An experimental and theoretical study of the transient régime of drop formation in such a system is the aim of this paper.

2. Experiments

The experiments were performed for the system of two immiscible liquids of very close densities: a solution of castor oil and dibutyl phthalathe was chosen for the dispersed phase and tap water for the continuous one. The physical properties and the test conditions are listed in Table 1 (where lower index d denoted the dispersed phase and c the continuous one).

Density	Density	Viscos	ity	Interfacial tension	Mean velocity
Qd,c	Qa-Qc	μ _d	μ _c	σ	through orifice U
g/cm ³	g/cm ³	Poi	ses	dynes/cm	cm/s
1.0	10-3-5.10-3	1.0	0.01	15-20	1-15

Table 1

The internal orifice diameter D was equal to 0.1977 cm. This diameter represented a characteristic length. The group of factors controlling the phenomenon of drop formation includes: densities of liquids ϱ_d and ϱ_c , their viscosities μ_d and μ_c , interfacial tension σ , diameter of the orifice D and mean velocity of efflux U. Taking into consideration the difference between densities $\Delta \varrho$ instead of density ϱ_c and gravitational acceleration g we can obtain by means of the dimensional analysis the system of five non-dimensional numbers determining the similarity in these experimental conditions:

(2.1) Bo
$$= \frac{\Delta \varrho g D^2}{\sigma}$$
, We $= \frac{\varrho_d U^2 D}{\sigma}$, Re $= \frac{\varrho_d U D}{\mu_d}$,
 $\overline{\mu} = \frac{\mu_d}{\mu_c}$, $\overline{\varrho} = \frac{\Delta \varrho}{\rho_d}$.

These numbers show the range of experiments in Table 2.

Table 2

Во	We	Re	μ	ē	
.0025—0.01	0.015-3.0	0.2-3.0	100	10-3-5.10-3	

The measurements and observations of phenomena were mainly recorded photographically and the mean values of resulting variables (like drop sizes, mean flow rates, etc.) were determined by the weight method.

3. Experimental results and theoretical models

The typical pattern of drop formation in the transient régime of a dispersion can be outlined as follows: the jet that remains after the detachment of the former drop contracts to the orifice (Fig. 2a) and there it forms a spherical inception of the next drop (Fig. 2b).

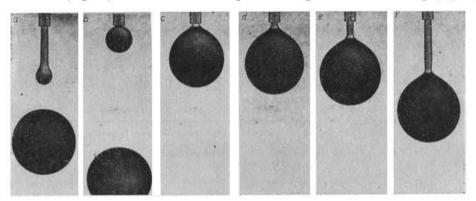


FIG. 2. Drop formation in the transient régime of dispersion.

The drop remains at the edge of the orifice and it grows in volume filled from the orifice (Fig. 2c) up to the moment when it begins to move at the end of a jet (Fig. 2d). This motion is continued (Fig. 2e) to the point of drop detachment (Fig. 2f).

One can observe the characteristic changes of the distance measured from the top of a drop to the plane of the orifice in the time of this period. This distance (denoted by h) was chosen as the one of the main variables which describes the phenomenon. An exemplary relation for this distance as a function of time taken from experimental movie-pictures is drawn in Fig. 3. On this diagram it is easy to point three characteristic stages of drop formation. In the first stage, the jet contracts to the orifice, in the second one, the drop

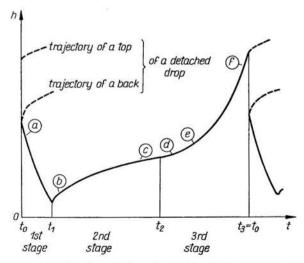


FIG. 3. Height of a drop as a function of time. (Letters in circles correspond to photos in Fig. 2).

remains on the orifice, and in the third one, it travels at the top of a jet up to the moment of detachment. If the velocity of the efflux U is increased and if it is greater than a certain value U_j called the jetting point [6], then the description of drop formation should be modified [2, 21]. In this situation the drop that forms in the way of jet contraction in the 1st stage does not reach the orifice. In the 2nd stage, the short jet exists between the nozzle and a growing drop. Further increases of the velocity U shorten this stage up till it disappears completely. The range of velocities mentioned above belongs to the jet régime of a dispersion.

Some simple mathematical models were set up to allow for a physical interpretation of the phenomena observed in a transient régime. In the 1st stage and the 3rd one of drop formation the motion of the drop at the end of a jet seems to be a main factor of the phenomenon. Consequently, the mathematical models for these stages are based on the law of drop momentum conservation. The drop will be treated as an individual spherical body (resulting from small Bond number conditions) which can interchange the mass and the momentum with a jet. The outer factors (as a jet and the outer liquid) act upon the drop by a system of forces. We take into account those forces which are due to interfacial

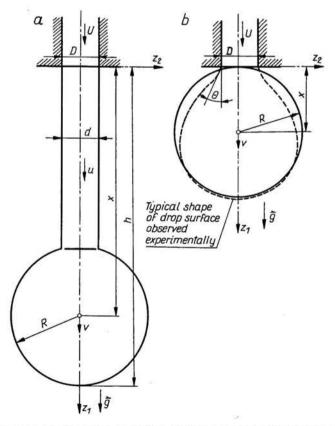


FIG. 4. Configuration for the theoretical description of drop formation: a) in the 1st stage and the 3rd stage, b) in the 2nd stage of formation.

tension, gravitation and stresses at the liquid-liquid interface. The definitions of these forces result from additional assumptions.

We assume that the system of forces acts upon the drop in such a way that the mass center of a drop moves along a straight line that coincides with the axis of symmetry of the orifice. It is chosen as one of the axes of the motionless coordinate system fixed with the orifice — that one which conforms to the direction of gravitational acceleration \bar{g} (Fig. 4).

The mass of a drop m and the velocity of its mass center v will be determined as the functions of time t from the system of equations consisting of a one-dimensional equation of motion of a drop mass-center (based on the law of momentum conservation for the material point of variable mass [24] p. 134) and the relation for the rate of increase of drop mass.

If the mean velocity in a jet (in the cross-section that connects the jet and the drop) is u and the diameter of the jet is d, then the system mentioned has a general form:

(3.1)
$$\frac{d}{dt}(mv) = \sum_{k} F_{k} + \frac{dm}{dt}u,$$
$$\frac{dm}{dt} = \varrho_{d} \frac{\pi d^{2}}{4}(u-v).$$

The determination of the forces F_k acting upon the drop is based on the assumption that the system of stresses on the drop surface looks like in the Stokes' motion of an individual liquid sphere moving with constant velocity through another liquid [25]. Additional forces are due to gravitation and interfacial tension. The system of forces has the following form:

$$F_i = -\pi d\sigma \cos\theta$$
, due to interfacial tension,
 $F_g = mg$, raising from gravity,

(3.2) $F_v = -\left(\frac{6\pi^2}{\varrho_d}\right)^{1/3} \mu_c \frac{2\mu_c + 3\mu_d}{\mu_c + \mu_d} vm^{1/3}$, as a viscid force according to Hadamard-

Rybczyński's law [25],

$$F_p = -\varrho_c \frac{m}{\varrho_d}g + \frac{\pi d^2}{4}\sigma K.$$

In the definition of the last force F_p , the first term results from the pressure distribution on the drop surface, and the second one follows from the additional pressure which exists in the cross-section connecting the jet and a drop [26]. This pressure is due to interfacial tension and equals to σK , where K is the curvature of the interface in the section considered. Due to the fact that it depends on the local shape (changeable) of a drop-jet connexion, the determination of this pressure is rather difficult to do. When the drop radius is close to the jet diameter, then it seems reasonable that this curvature is close to the curvature of the jet (K = 2/D if the jet is cylindrical). We assume that in other cases the value of the curvature is the same as the one above.

The parameters describing the jet, namely its diameter d and the mean velocity u, are in general cases also unknown functions of time. Since the liquid flow rate q in a jet can be assumed to be constant, then:

$$(3.3) q = \frac{\pi d^2}{4} u,$$

and the system (3.1)+(3.3) ought to be completed by one more equation. Instead of that we assume the following, what simplifies the problem:

$$(3.4) u = U = \text{const}, \quad d = D = \text{const}.$$

As it was mentioned above, the height of a drop h can be chosen as a convenient quantity in examining the results of the experiments. Now we define it by:

$$(3.5) h = x + R,$$

where x is a coordinate of a drop mass center and can be obtained from a familiar equation:

$$\frac{dx}{dt} = v,$$

and R is the drop radius defined due to drop sphericity by:

(3.7)
$$R = \left(\frac{3m(t)}{4\pi\varrho_d}\right)^{1/3}.$$

The equation (3.6) and the system (3.1) will be solved together. Substituting (3.2) and (3.4) into (3.1) and then introducing the following dimensionless quantities

(3.8)
$$\overline{m} = \frac{m}{\varrho_d \pi D^3}, \quad \overline{v} = \frac{v}{U}, \quad \overline{x} = \frac{x}{D}, \quad \overline{t} = \frac{tU}{D},$$
$$\overline{R} = \frac{R}{D} = \left(\frac{3}{4}\overline{m}\right)^{1/3}, \quad \overline{h} = \frac{h}{D}$$

we can obtain the non-dimensional form of the system (3.1)+(3.6):

(3.9)
$$\frac{d}{d\bar{t}}(\bar{m}\cdot\bar{v}) = \frac{Bo}{We}\bar{m} - \frac{b}{We} - 6^{1/3}\frac{(2+3\bar{\mu})\bar{v}\bar{m}^{1/3}}{\bar{\mu}(1+\bar{\mu})Re} + \frac{d\bar{m}}{d\bar{t}},$$
$$\frac{d\bar{m}}{d\bar{t}} = \frac{1}{4}(1-\bar{v}), \quad \frac{d\bar{x}}{d\bar{t}} = \bar{v},$$

where Bo, We, Re, $\overline{\mu}$ are criterial numbers of similarity defined by (2.1) in the description of experiments and b is a coefficient defined by

$$(3.10) b = \cos\theta - \frac{1}{2}.$$

For every stage of drop formation we, in turn, determine the conditions for the beginning and the end of the stage. It is assumed that the drop mass \overline{m} and the coordinate \overline{x} of its center are continuous functions of time \overline{t} but the velocity \overline{v} may not be continuous from stage to stage.

Considering the 1st stage we assume additionally that viscid force F_v can be neglected and that

$$(3.11) \qquad \qquad \cos\theta = 1$$

because of the assumed cylindrical shape of a jet.

The condition for the beginning of this stage is specified as

(3.12)
$$\bar{t} = \bar{t}_0$$
: $\bar{m} = 0$, $\bar{v} = 1 - \sqrt{\frac{2}{We}}$, $\bar{x} = \bar{x}_0$,

where the coordinate of the mass center \overline{x}_0 is determined from the length of a jet remaining after drop detachment. The values of \overline{x}_0 in calculations are introduced from data of the experiments. The system (3.9) simplified in such a way has an exact solution of the form:

(3.13)

$$\overline{m} = \frac{1}{2} \left(\frac{\overline{t} - \overline{t_0}}{\sqrt{2We}} + \frac{Bo}{12We} (\overline{t} - \overline{t_0})^2 \right),$$

$$\overline{v} = 1 - \sqrt{\frac{2}{We}} + \frac{1}{3} \frac{Bo}{We} (\overline{t} - \overline{t_0}),$$

$$\overline{x} = \overline{x_0} + \left(1 - \sqrt{\frac{2}{We}} \right) (\overline{t} - \overline{t_0}) + \frac{1}{6} \frac{Bo}{We} (\overline{t} - \overline{t_0})^2.$$

The end of this stage is determined by the moment when the drop reaches the plane of the orifice (Fig. 2b) and hence the condition in this theoretical description is:

$$(3.14) \qquad \qquad \overline{t} = \overline{t}_1 > \overline{t}_0; \qquad \overline{x} = \overline{R},$$

where \overline{R} (dimensionless radius of spherical drop) is defined by

$$(3.15) \qquad \qquad \overline{R} = \left(\frac{3}{4} \ \overline{m}\right)^{1/3}.$$

For the moment determined by (3.14) we can find from (3.13) the mass \overline{m}_1 and the period \overline{t}_1 of the 1st stage. This mass will be applied in the initial condition of the 2nd stage.

It should be noted that this simple solution (3.13) admits three cases presented schematically in Fig. 5:

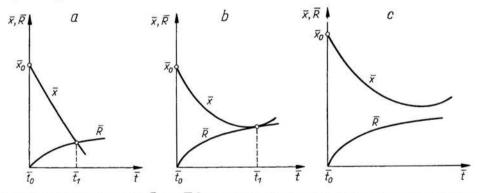


FIG. 5. Graphs of relations for $\overline{x}(t)$ and $\overline{R}(t)$ predicted by the theory in the 1st stage of formation when: a) $U < U_j$, b) $U = U_j$, c) $U > U_j$.

- a) where graphs of $\overline{x}(t)$ and R(t) intersect,
- b) where the curves are tangent in the point $t = t_1$,
- c) where there is no common point.

All these cases were observed in experiments. The situation from Fig. 5b determines in this mathematical model the jetting point when $U = U_j$. If U is greater than U_j (Fig. 5c), the jet does not contract up to the plane of an orifice and the jet régime of dispersion is reached.

Considering the case from Fig. 5b where the following condition is satisfied:

(3.16)
$$\overline{t} = \overline{t}_1, \quad \overline{x} = \overline{R}, \quad \frac{dR}{d\overline{t}} = \overline{v},$$

the upper limit of the range of the transient régime can be estimated through this theoretical approach. We noted a slight disagreement between theoretical and experimental values of the velocity U_i (experimental values are a little higher than theoretical ones).

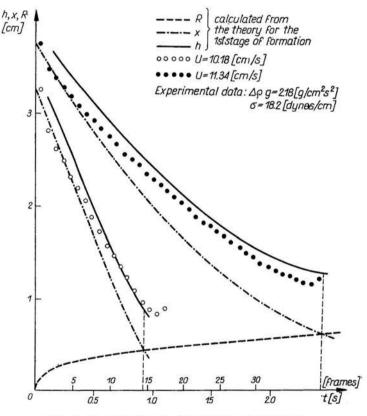


FIG. 6. Jet contraction in the 1st stage of formation.

The diagram in Fig. 6 shows the results of both calculated and measured (from movie pictures) heights of the drop h plotted against the time t in the first stage of formation.

In the 2nd stage, observations show that the shapes of the drop pendant on the orifice are very close to those described by the statics [27]. Nevertheless, static stability solutions [27, 28] seem to be inapplicable to determine the values of the mass of the drop which begins to travel from the orifice (i.e. the final mass of the second stage \overline{m}_2). We assume that in this stage the spherical drop remains tangent to the plane of the orifice

and hence the velocity of its mass center is

(3.18)
$$\bar{v} = \frac{dR}{d\bar{t}} = \frac{1}{4} (6\bar{m})^{-2/3},$$

because the equation for the mass increase is

$$\frac{d\overline{m}}{d\overline{t}} = \frac{1}{4},$$

i.e. the whole liquid flowing out of the orifice comes into a drop. For $\overline{t} = \overline{t}_1$ is $\overline{m} = \overline{m}_1$ and from (3.19) we get the linear function:

(3.20)
$$\overline{m} = \overline{m}_1 + \frac{1}{4} (\overline{t} - \overline{t}_1).$$

In order to specify the condition for the end of the 2nd stage it must be noted that when the drop grows hanging on the edge of the orifice the variation of the angle θ (Fig. 4b) is physically admissible due to the 90° angle of the edge of the orifice. We shall suppose that the changes of θ in definition (3.2) of the force F_i resulting from interfacial tension have such local importance that they are not contradictory with the assumed spherical shape of a drop. Next, it is assumed that the angle θ is changeable in such a way that the first equation of the system (3.9) with the neglected viscid term is satisfied. Hence, the relation for $\cos\theta$ can be written in the form:

(3.21)
$$\cos\theta = \operatorname{Bo} \cdot \overline{m} + \frac{\operatorname{We}}{4} \left(1 - \frac{1}{3} \overline{v} \right),$$

where \overline{v} and \overline{m} are already determined by (3.18) and (3.20). The end of the 2nd stage is defined by the moment of the beginning of jet creation. The assumption for the shape of the jet (3.4) yields the following condition for the moment

Substituting it into (3.21) we can find the mass \overline{m}_2 and then $\overline{x}_2 = R_2$ [definition (3.15)].

The description of drop formation in the 3rd stage is given by the full system (3.9) and the initial condition:

(3.23)
$$\overline{t} = \overline{t_2}; \ \overline{m} = \overline{m_2}, \ \overline{v} = 0, \ \overline{x} = \overline{x_2},$$

where we put $\overline{v} = 0$ because in this stage we will neglect the velocity of the spherical growing of the drop in favour of the velocity of its motion at the end of a jet.

The important and difficult problem is the determination of the end of the 3rd stage that is physically indicated by the end of the process of the detachment of a drop. The theoretical model presented here does not allow for a mathematical description of this process. The direct conclusion obtained from the introduced condition for the beginning of the 1st stage is:

$$(3.24) \qquad \qquad \overline{t} = \overline{t_3} \colon \overline{x_3} = \overline{x_0} + \overline{R_3}$$

as the condition for the end of integrating the system (3.9) and the end of the period of drop formation. In this way we obtain from (3.9) the final mass of the drop \overline{m}_3 that tears off the jet.

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The exemplary relations for $\overline{m}(t)$, $\overline{v}(t)$ and $\overline{x}(t)$ in the whole period of formation predicted by the presented theoretical approach in the transient régime are illustrated in Fig. 7. The results of theoretical calculations in the form of a relation between the height

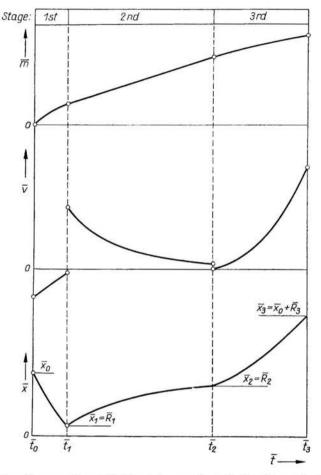


FIG. 7. Typical relations for mass \overline{m} , velocity \overline{v} and the coordinate \overline{x} of a drop as the functions of time t predicted by the theoretical approach.

 \overline{h} and the total volume \overline{V} of a drop and a jet are compared with the results of the experimental measurements in Fig. 8 for the experimental value of Bond number Bo = 0.0061. Considering the influence of the simplifying assumption of drop sphericity it is of interest to note that the agreement between theoretical and experimental (smoothed) curves seems to be reasonable in the 1st and the 2nd stages. In the 3rd stage of drop formation during which a relatively great drop is in motion at the end of a short jet, the picture indicates that the resistance of drop motion is greater than it is predicted by the theoretical model. Moreover, it leads to the fact that the masses \overline{m}_3 obtained from calculations are smaller than the respective experimental values (Fig. 9).

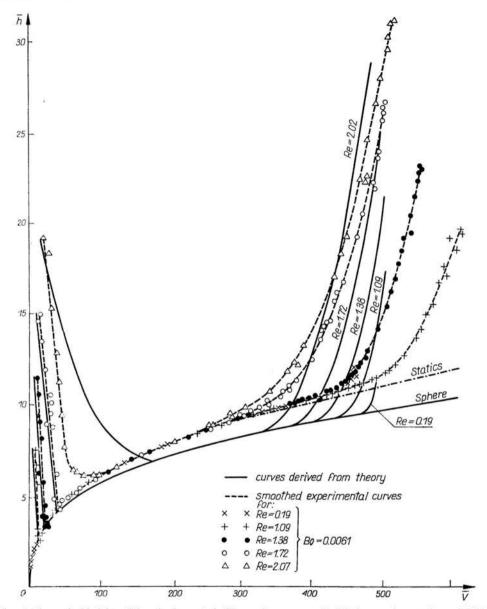


Fig. 8. Theoretical heights of drop in the period of formation compared with the experiments (Bo = 0.0061).

4. Concluding remarks

The experimental study of drop formation was performed in a system where one liquid flows out of the orifice into another immiscible liquid. The liquids of the system were chosen in such a way that they had almost the same density. This corresponds to the very small Bond numbers and has the advantage that the experimental procedure is greatly

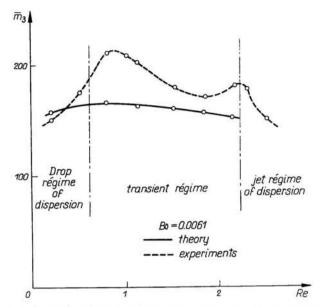


FIG. 9. Calculated and measured values of drop mass after detachment.

simplified. This advantage is considerable in a study concerning the transient régime of dispersion where little information is available. The process of drop formation in this régime is described in the paper by a simple theory that exploits the characteristic features of the process observed experimentally. The agreement between the experimental and the theoretically predicted course of formation was found to be reasonable, considering the simplicity of the theoretical approach. It can be said that in order to obtain better agreement the assumptions in theory could be modified in some directions:

the condition of constant velocity in a jet may be replaced by a model of flow in a jet, the spherical shape of a drop can be modified to a shape derived e.g. from statics, etc.

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