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THE MARANGONI EFFECT AND TRANSLATION OF FREE NON-DEFORMABLE DROPS

E. Chifu, I. Stan, Z. Finta, and E. Gavrila

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THE MARANGONI EFFECT AND TRANSLATION OF FREE NON-DEFORMABLE DROPS

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Drops suspended in an immiscible liquid undergo a complex motion $\frac{765}{7}$ when an interface tension gradient appears at the surface. In this context, studies can be mentioned on the pulsations of suspended drops [1-5] and the "interface activity" of "free" drops [6-7].

Recently, chemical and hydrodynamic instability has been studied for fluid interfaces, instabilities of the Prigogine-Glansdorff "disipative structure" type, in which interface tension is the parameter linking chemical and hydrodynamic processes [8-21].

The phenomena generated by interface tension gradients have been arousing special interest in recent years in the problem of mass transfer at fluid interfaces [19-20] and in modelling certain processes of deformation and motion at the biosurface level [8-11, 14, 21]. Finally, because surface (interface) tension plays a decisive role in the behavior of liquids at zero gravity [22-30], the authors consider that the phenomena defined by gradients of this size will also occur in space.

In this work, phenomena are studied, theoretically and experimentally, which are caused by interface tension gradients in the case of "free" drops. Such a drop, devoid of buoyancy (motion), is obtained by suspending it in a liquid of the same density. The studies carried out shew that, as a primary result of interface tension gradient, a <u>sur-</u> "Numbers in the margin indicate pagination in the foreign text.

face flow or so-called <u>Marangoni effect</u> is produced at the drop surface [31-42]. Upon the action of surface-flow processes, a <u>translation</u> <u>motion</u> then appears in the drops as a whole, although the initial drop was immobile.

Model of Surface Flow and Translation Motion of Free Drops

Let us consider drop L', with radius \underline{a} , suspended in an immiscible liquid L (Fig. 1). Force \vec{F} , which acts as a unit drop volume: $\vec{F} = (\rho' = \rho)\vec{r}$

will go to zero, when the densities of these two liquids are equal $(\rho' = \rho)$, or in zero gravity $(\vec{r} = 0)$.

Such a "free" drop and initial lack of motion (buoyancy) are assumed.

Let us assume that the initial liquid/liquid interface tension is σ_0 . At point P_1 (Fig. 1) on the drop surface, the interface tension is then reduced (σ_1) , by injecting a surfactant, while at the "pole" opposite P_0 , it remains as initially (σ_0) . Along the meridian of the $\frac{760}{760}$ drop, a gradient (difference) appears for interface tension $(\sigma_0 - \sigma_1)$.

The processes which appear as a result of this gradient are complex, and it is probable that at least the following must be taken into consideration:

- displacement of drop translation and its eventual rotation;

- deformation and oscillation of the drop, appearance of waves on its surface;

- circulatory currents in the liquid.

In revealing such processes, Valentine and co-authors [6-7]estimate the order of size for energy dissipated in different forms of drop "motion", but they consider that solving the equations of fluid motion is a much too complicated problem.

It is noted that up to now no rigorous and complete theory has existed for the phenomena generated for interface tension gradient relative to a "free" drop.

In an effort to solve the problem, the authors of the present study consider that the Marangoni effect, relative to surface flow [31-43], must be taken into account first of all in a quantitative treatment. It is surprising that the authors cited [6-7] do not expressly specify this process among the phenomena enumerated, although it represents the most direct effect of interface tension gradient.

From Figure 1, it results that, upon the action of the gradient $(\sigma_0 - \sigma_1)$, flow takes place at the drop surface itself L'. Simultaneously, one layer of the liquid L, which corresponds to the arrows on the outside of the sphere, undergoes movement, and liquid currents appear in the interior of the drop L' (dotted arrows). These last two processes are explained by the absence of slip between the surface proper of the drop, activated by the interface tension gradient, and the two liquids (L and L').

Due to surface flow involving the exterior of the liquid L, forces of hydrodynamic pressure act upon the drop L'. Their resultant must define the translation motion of the whole drop in the direction which links the points of the drop's maximum and minimum interface tension (Fig. 1, arrow pointing straight up).

Experimental studies have led the authors to the conclusion that two limiting cases exist:

- large interface-tension gradients and reduced drop viscosity, in which situation the drop is highly deformed, oscillation phenomena appear, as well as its possible fission, etc.;

- low interface-tension gradients and elevated drop viscosity, in which situation the drop behaves as being practically non-deform able, like a rigid sphere.

In this work, the latter case is studied. Using non-deformable drops has permitted the direct experimental determination of the rate of <u>surface flow</u>. At the same time, it has been possible to "isolate", at least partially, <u>translation motion</u> of the whole drop from the other effects (deformation, oscillation, rupture, etc.) which appear as a result of interface tension gradient.

Utilization of a non-deformable drop model has led, in addition, to a satisfactory theoretical treatment of the hydrodynamics of these two processes.

Hydrodynamics of the Marangoni Effect at a Spherical Fluid Surface

Let us assume that:

- the liquids L and L' are viscous and incompressible;

- the densities (p, p'), viscosities $(v = \mu/p, v' = \mu'/p')$, and temperatures (T, T') are constant;

- the volume force $(\rho' - \rho)\vec{g}$ which acts upon the drop is zero, according to the assumption in the model described;

- flow caused by the interface tension gradient is, in the first approximation, stationary;

- the Reynolds number (R) is small;

-

- the Bond (Bo) and Weber (We) numbers are less than one, and the forces of acceleration and inertia are small in comparison with those of surface tension, in conformance with the model proposed; this implies that interface tension <u>g</u> is dominant in determining the flow of the liquids [22].

The equations which govern flow are continuity equations:

$$\operatorname{div} \vec{v} = 0 \tag{1}$$

$$\operatorname{div} \vec{v} = 0 \tag{2}$$

where $\frac{1}{r}$ is the rate of flow of the medium (L), and \vec{r} for the liquid L' from the interior of the drop, and the Navier-Stokes equations are:

$$(\vec{v} \nabla) \vec{v} = -\frac{1}{\rho} \operatorname{grad} p - v \bigtriangleup \vec{v}$$

$$(3)$$

$$(\vec{v} \nabla) \vec{v} = -\frac{1}{\rho} \operatorname{grad} p - v \bigtriangleup \vec{v}$$

$$(4)$$

p and p' being the pressures at the exterior and interior of the drop. Because for a small Reynolds number, convection terms can be neglected, equations (3) and (4) become:

grad
$$p = \mu \Delta \vec{v}$$

grad $p' = \mu \Delta \vec{v}$ (6)

The symmetry of the problem suggests a spherical coordinate sys- $\frac{767}{167}$ tem (r, 0.9.) with the origin at the center of the drop and the Oz axis intersecting the sphere at the point of minimum interface tension σ_1 (Fig. 2). Assuming that motion is symmetric with respect to the Oz axis, the liquid flow rates for exterior L and interior L' are in-

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(5)

dependent of angle φ ; these rates will have only a normal (radial) component;

$$v_r(r, \theta); = v_r(r, \theta)$$

and a tangential one, at the drop surface:

$$v_{\theta}(r, \theta); v_{\theta}(r, \theta)$$

In spherical coordinates, equation (1) becomes [43]:

$$\frac{\partial v_r}{\partial r} = \frac{1}{r} \frac{\partial v_\theta}{\partial r} = \frac{2v_r}{r} - \frac{v_\theta \operatorname{ctg} \theta}{r} = 0, \qquad (7)$$

and equation (5) is written as:

$$\frac{\partial p}{\partial r} = \mu \left(\frac{\partial^2 v_r}{\partial r^2} - \frac{1}{r^2} \frac{\partial^2 v_r}{\partial \theta^2} - \frac{2}{r} \frac{\partial v_r}{\partial r} - \frac{\operatorname{ctg} \theta}{r^2} \frac{\partial v_r}{\partial \theta} - \frac{1}{r^2} \frac{\partial^2 v_r}{\partial \theta} - \frac{2}{r^2} \frac{\partial v_{\theta}}{r^2} - \frac{2}{r^2} \frac{\operatorname{ctg} \theta}{r^2} v_{\theta} \right),$$

$$\frac{1}{r} \frac{\partial p}{\partial \theta} = \mu \left(\frac{\partial^2 v_{\theta}}{\partial r^2} - \frac{1}{r^2} \frac{\partial^2 v_{\theta}}{\partial \theta^2} - \frac{2}{r} \frac{\partial v_{\theta}}{\partial r} - \frac{\operatorname{ctg} \theta}{r^2} \frac{\partial v_{\theta}}{\partial \theta} - \frac$$

The equations for liquid motion at the drop interior have a similar form.

The components of the rates at the exterior L and interior L' of the liquids must satisfy the following conditions:

- the normal components must be zero at the surface of the nondeformable drop:

$$\mathbf{t}_r = \mathbf{t}_r' = \mathbf{0} \tag{10}$$

- the tangential components must be equal at the drop surface (continuity):

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 $v_0 = v_0' \quad \text{at} \quad r = a, \tag{11}$

- the rate at the interior of the drop must be finite at every point, particularly at its center:

$$v_{i}$$
 is finite for $v = 0.$ (12)

and goes to zero away from the drop:

$$v_r = v_0 = 0 \quad \text{for} \quad r = \infty \tag{13}$$

In addition to these kinematic conditions, it is necessary for a dynamic condition to be fulfilled: the condition of continuity of the tangential components p_{i0} and p'_{i0} for the tension tensors and the gradient of interface tension $\vec{p}_t = \operatorname{grad} \sigma$ [43]:

$$Pro - Pr = Pro$$

In spherical coordinates, the condition can be written thus:

$$\mu \left(\frac{1}{r} \frac{\partial v_r}{\partial \theta} - \frac{\partial v_{\theta}}{\partial r} - \frac{v_{\theta}}{r} \right)_{r=a} - \frac{1}{a} \frac{\partial \sigma}{\partial \theta} = -\mu' \left(\frac{1}{r} \frac{\partial v'_{\theta}}{\partial \theta} - \frac{\partial v'_{\theta}}{\partial r} - \frac{v'_{\theta}}{r} \right)_{r=a}$$
(14)

The solution for equations(7-9) for flow of the liquid exterior L is, taking conditions (10-14) into account:

$$v_r = \frac{(\sigma_0 - \sigma_1)a^2}{6(\mu + \mu')} \left(\frac{1}{r^2} - \frac{1}{a^2r}\right) \cos \theta, \qquad (15)$$

$$t_{\theta} = \frac{(\sigma_{\theta} - \sigma_{1})a^{2}}{6(\mu + \mu')} \left(\frac{1}{2r^{2}} + \frac{1}{2a^{2}r}\right) \sin \theta, \qquad (16)$$

$$P = -\frac{\mu (\sigma_{0} - \sigma_{1}) a}{6 r^{2} (\mu + \mu')} \cos \theta$$
(17)

while for the liquid L' at the interior of the drop, we obtain;

$$v'_{r} = \frac{(\sigma_{0} - \sigma_{1})}{6(\mu - \mu')} \left(1 - \frac{r^{2}}{a^{2}}\right) \cos \theta.$$
 (18)

$$r_{\theta}' = -\frac{(\sigma_{\theta} - \sigma_{1})}{6_{e}(\mu - \mu')} \left(1 - \frac{2r^{2}}{a^{2}}\right) \sin \theta,$$
(19)
$$p' = -\frac{5}{3} \frac{\mu'(\sigma_{\theta} - \sigma_{1})r}{a^{2}(\mu + \mu')} \cos \theta$$
(20)

The liquid flows along the surface of the drop, where $v_0 = v'_0$, conforms to condition (11), with the rate;

$$(r_{\theta})_{r=\theta} = \frac{\sigma_{\theta} - \sigma_{1}}{6(\mu - \mu)} \sin \theta = r_{\theta} \sin \theta,$$
 (21)

which results alternatively from equations (16) and (19) for r = a. The value:

$$v_0 = \frac{\sigma_0 - \sigma_1}{6(\mu + \mu')}$$
 (22)

represents the maximum rate found at the drop equator.

As for flow at the surface itself of the drop, a term could be introduced corresponding to surface viscosity. The calculations done by the authors, which will be adopted again in further works, show that surface viscosity of expansion [33] appears in equations (15-22), as a correction term, which - for the systems studied - can be neglected in the first approximation.

Finally, the phenomena of adsorption have not been taken into <u>/768</u> consideration, nor have those of diffusion, because it is assumed that these processes are sufficiently rapid in comparison with surface flow. Interference of these phenomena with the Marangoni effect at a spherical fluid surface will be the subject of future research.

Experimental testing of the hydrodynamic model proposed, relative to equations (21-22), have led to conclusions which are totally satisfactory.

Measurements of Surface Flow. Discussion

The experimental study of surface flow was done on a liquid/ liquid system with equal densities; several of the systems studied, with density $P = P' = 0.863 \text{ g/cm}^3$ are presented in Table 1.

All the measurements were made at a temperature of $20 \pm 0.1^{\circ}$ C.

The continuous phase L was placed in a rectangular vessel with a capacity of 1 dm^3 , with transparent walls. A drop of L' was pipetted into the continuous phase L, and the final density was then adjusted by small additions of water or alcohol, until the buoyancy of the drop practically disappeared.

After stabilization of the system, $10^{-3}-10^{-2}$ cm³ of a solution of a surface-active agent was injected at a point on the surface of the drop (P₁, Fig. 1). With the aim of making surface flow visible, the surfactant solution was strongly colored with methylene blue (0.28 g/100 cm³). The frontal advance of the surfactant was tracked by means of cinematography with a high-speed camera (500 images/sec). With the aid of a simple trigonometric relationship, from a blown-up projection of the filmed images, the distance 1 was determined which was covered by the front along the meridians of the drop at different instants t, the number of the images being in direct correlation with time. The rate of surface flow $(r_0)_{res}$ is given by the derivative of the curve I = f(t).

In Figure 3, the curves l=f(t) are represented for some of the systems studied. The shape of these curves, usually an 5, indicates /769 the qualititive agreement between experiment and theory, the maximum value of the rate, in conformance with equation (21), being attained

at the equator of the drop and marked with an arrow on Fig. 3.

Pronounced dispersion of the data, caused by various experimental difficulties, did not permit sufficiently precise evaluation of the flow rate over the entire surface of the drop. Nevertheless, the average volues of f_{mexp} corresponding to the equator, for drops of different radius, were in satisfactory agreement with those calculated f_{mexp} according to equation (22), such as result from Table 2 (the systems from Table 1).

The results contained in Table 2 generally reflect well the role of the interface tension gradient and the viscosity of the liquids in surface flow.

Taking into account the values for the rates of flow and other parameters from Table 1, the Reynolds numbers $R = \frac{2}{A} \frac{2}{P} \frac{r_{0,exp}}{(\mu + \mu')}$ are close to unity; that is, the assumptions which were made for the hydrodynamic model are correct.

In many cases (systems 2-4), the experimental values for flow rate are larger (by 20%) in comparison with the theoretical ones, which is, at first glance, surprising. It is considered that this deviation is explained by the fact that the drop acted upon by an interface tension gradient is not immobile, as is assumed in the hydrodynamic Marangoni effect, but undergoes translation motion as a consequence of the very processes of surface flow (see the model in Fig. 1). This effect is studied below.

Translation Motion of Free Drops. Hydrodynamic Treatment

As is noted in the case of the experiments on surface flow which were carried out, the Reynolds numbers were close to unity and a viscous

flow regime exists. In turn, the experimental determinations of the rate of translation for whole drops led to values for this criterion in the range of 10-60. Due to this, in order to calculate the translation rate of a drop, one no longer has recourse to direct integration of the hydrodynamic equations; a different method is used.

Actually, according to the model for surface flow, involving the exterior liquid L (Fig. 1), forces of hydrodynamic pressure act on the drop L'. Taking into account the symmetry of surface flow, the resultant F_p of these forces is oriented in the positive sense along the axis Oz (Fig. 2). The force of propulsion F_p thus defines the translation motion of the drop as a whole, and its center of mass along the positive Cz axis.

The force F_p which appears due to flow of the liquid L around the drop L' have the form [44]:

$$F_p = \iint_{S} (p_{r_2} \cos \theta - p_{r_0} \sin \theta) \, ds. \qquad (23)$$

where p_{cr} and p_{ce} are the normal and tangential components, respectively, of the tensor of viscous tension:

$$p_{rr} = -p + 2\mu \frac{\delta v_r}{\delta r}, \qquad (24)$$

$$p_{14} = \left(\begin{array}{ccc} 1 & \partial_{1r} & \partial_{10} & 10 \\ r & \partial_{1} & \partial_{1} & r \end{array} \right)$$
(25)

Since the element of the spherical-drop surface is:

$$ds = 2 \ \pi a^2 \ \sin \theta \ d\theta$$

equation (23) is transcribed as:

$$F_{p} = \int_{0}^{\pi} (p_{rr} \cos \theta - p_{rg} \sin \theta) 2\pi z^{2} \sin \theta d\theta, \qquad (26)$$

But, at the drop surface, for $r_{=a}$, we have $r_{r}=0$. according to equation (15), and, in addition:

$$\frac{\partial r}{\partial r} = \frac{ur}{\partial t} = 0 \tag{27}$$

Thus, in order to obtain a normal (radial component or the tension tensor, taking into account equations (24), (27), and (17);

$$p_{rr} = \frac{\mu(\sigma_0 - \sigma_1)}{6 a(\mu - \mu')} \cos \theta, \qquad (28)$$

The tangential component of the tensor is, in conformance with equations (25), (27), and (16):

$$p_{r_{\theta}} = -\frac{\mu}{2} \frac{(\sigma_{\theta} - \sigma_{1})}{a(\mu + \mu')} \sin \theta. \qquad (29)$$

Finally, introducing the values and from equations (28) and (29) into equation (26), the following expression is obtained by integration for the resultant force F_p :

$$F_{p} = \frac{14 \pi \mu a (\sigma_{0} - \sigma_{1})}{9 (\mu - \mu')}$$
(30)

The force F_p of propulsion for the entire drop acts as a motive force, which is directly proportional to the difference in interface tension, causing translation of the drop.

The force of resistance F_r which opposes the displacement of a spherical drop takes the form [43, 45]:

$$F_r = \frac{1}{2} \pi a^2 \varphi u^2 C_d \tag{31}$$

where u is the rate of displacement, while C_d is the coefficient of resistence, which depends on the Reynolds number. For Reynolds numbers lower than 300, the coefficient C_d for a liquid drop is the same as for a solid sphere with identical radius and can be obtained by interpolation from the curve $R = R(C_d)$ [45].

By making the forces from equations (30) and (31) equal, .e rate /770 u is obtained at which the translation displacement of the entire drop must take place:

$$u = \sqrt{\frac{28}{9} \frac{\mu(\sigma_{0} - \sigma_{1})}{\rho \sigma C_{d} (\mu + \mu')}}$$
(32)

It is evident that equation (32) describes only the initial rate of drop displacement. Its motion is damped gradually, however, due to the disappearance of the interface tension gradient, by the process of surface flow itself, and as a result of friction with the viscous medium. The entire phenomenon has a transitory character.

The fact must be emphasized that, in the hydrodynamic model proposed, it is assumed that the translation of entire "free" or "weightless" drops (initially without motion), at the surface of which interface tension gradients act, is explained by the processes of surface flow. This fact has not been described as yet in the literature.

Experimental Determination of the Rate of Translation for Free Drops. Discussion

Evaluation of the rate of translation for whole drops - essentially non-deformable - was done in the same system (Table 1) in which the rate of surface flow was determined.

The experiments were conducted in a similar way to those for surface flow, with the distinction that for these data, translation of the drop as a whole was used:

- a "free" drop, initially at rest due to a lack of buoyancy, is injected with a surfactant solution at a point on the surface, interface tension here being σ_1 , while at the "pole" opposite to it, it is momentarily as initially, σ_0 (Fig. 1);

- as a result of the gradient of interface tension $(\sigma_0 - \sigma_1)$, a

surface flow appears (made visible by coloring the surfactant solution with methylene blue) and, after a certain "induction" period, translation of the drop is produced;

- displacement of drop translation, relative to a fixed reference point, resulted during filming at a speed of 24 images/sec; the distance z covered by the drop in time t was read with thin films projected at a specific size ratio.

It is necessary to emphasize, first of all, the fact that translation motion remains in accordance with that indicated in the theoretical model proposed in this work, that is, along the positive Oz axis (Fig. 2), if injection is carried out at the point on the surface of interface tension $\alpha_{\rm p}$. Reproducible results are obtained, in particular, when injection is done laterally, because the existence of a small density gradient along the vertical can dis upt the advance of the drop.

In Fig. 4, the results are recorded which were obtained for the function z = z(t) for some of the systems studied. It is observed that, after a certain time of injection (t = 0), the rate becomes maximum and constant, after which it tends to decrease, which is normal, considering the transitory character of the phenomenon; the maximum value of the derivative u = dz/dt (solid straight line) is taken as the rate of drop translation.

In Table 3, for the 3 systems from Figure 4, the average experimental values u_{exp} are reproduced for the rate of drop translation, in addition to the values u_{calc} which resulted from equation (32). Because the calculation of the u_{calc} values necessitates a knowledge

of the coefficient of resistence C_d , the Reynolds numbers $R = 2a\rho\mu_{exp}/\mu$, are evaluated first and then, with the aid of the diagrams $R = R(C_d)$, the values for C_d are obtained by interpolation [45].

It is observed that the experimental values are 20-30% lower than those calculated using equation (32). Although at present it is not possible to give a complete explanation, it can be suggested, in the case of visible, non-deformable drops, according to the author, that the dissipation of surface energy occurs in a form of motion other than translation.

It is, then, worth mentioning the fact that the experimental rates of drop translation, on the order of 0.2-0.3 cm/sec, explain the higher values for the experimental rates of surface flow, in comparison with those calculated (Table 2), because the sense of the drop's translatory motion is opposite to that of the surface flow (Fig. 1).

<u>Conclusions</u>

The translation motion which a free, non-deformable drop - suspended in a liquid of equal density - executes upon the action of gradients of interface tension, is explained on the basis of a model proposed in this work:

- The gradient of interface tension determines surface flow the Marangoni effect; this process takes place, first of all, at the very drop surface, at the rate given by equations (21)-(22).

- Due to the absence of slip between the surface proper and the liquids at the exterior and interior of the drop, surface flow is brought about rough the viscosity of both fluid phases, the compon-

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ents of the rate corresponding to those described in equations (15-16) and (18-19).

- As a consequence of surface flow involving the exterior liquid, the drop is subjected to forces of hydrodynamic pressure, the resultant of which (eq. 30) determines the translation motion of the entire drop. Propulsion of the drop takes place in the sense of the positive Oz axis (Fig. 2), passing through the point of minimum interface tension.

- Drop translation has a transitory character, the initial rate given by equation (32) diminishing gradually, due to the disappearance of the interface tension gradient through the processes themselves of surface flow and as a result of friction with the viscous medium.

The experiments performed on practically non-deformable drops have permitted direct measurements of both the processes of surface flow and translation motion of a drop, the data being in generally satisfactory agreement with the hydrodynamic theory proposed in this work.

The results presented contribute to a knowledge of the mechanism of "interface activity" of free drops, initially lacking motion, upon the action of gradients of interface tension. They attest to the fact that surface flow is an elemental phenomenon which determines the process of translation as a whole for free, non-deformable drops.



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Fig. 1. Model of surface flow and drop $\frac{766}{766}$ translation.



Fig. 2. System of spherical coordinates. /767

TABLE 1. COMPOSITION AND PARAMETERS OF LIQUID/LIQUID SYSTEMS /768						
System	Continuous phase, L	Drop, L'	Surfactant	6 6. , dyn/cm	ш. сР	14 7, CP
1	Ethan ol (78,6% vol.) Wa ter	Oil of paraffin	i-Propanol (75,95% vol.) Water	3.2±0,4	2,26	80
2	Ethanol (78,6%, vol.) Water	Oil of paraffin	n-Propanol (77,3% vol.) Water	4,4±0,4	2,26	80
3	Methanol (78% vel) Water	Oil of paraffin	1-Propanol (75,95% vol.) Water	5,5±0,4	1,33	80
4	Methanol (78% vol.) Water	Oil of paraffin	n-Propanol (77,3% vol.) Water	6,7 ±0,4	1,33	80





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System	'a (EXP)	o (calc)	
1 2 3 4	$0,63 \pm 0.15 \\1,12 \pm 0.08 \\1,37 \pm 0.24 \\1.45 \pm 0.33$	$0,65 \pm 0.08 \\ 0.89 \pm 0.08 \\ 1.11 \pm 0.08 \\ 1.37 \pm 0.08$	

TABLE 3. RATE OF DROP TRANSLATION, cm/sec

 System
 "exp
 "calc
 R
 Cd
 "calc:"exp

 1
 0,21
 0,31
 19,1
 3,2
 68

 2
 0,31
 0,40
 47,9
 1,7
 78

 3
 0,36
 0,44
 55,6
 1,7
 82



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