

DIMENSIONLESS ANALYSIS OF THE FREE DROP MOVEMENT AT LOW REYNOLDS NUMBER

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ABSTRACT. We report on the movement of a free drop at low Reynolds number, initially at rest, by methods of similarity theory and non-dimensional analysis. Because the drop is motionless, we have introduced as scale velocity, a characteristic velocity, U_c , and a new dimensionless number, Ch , which is related to Reynolds number. This permits us to evaluate the Marangoni force in the dimensionless form, as well as the deformation, the translational motion and even the break up of a free drop, under surfactants adsorption.

Keywords: *drop deformation, drop break up, surfactant adsorption, dimensionless number, Marangoni force.*

INTRODUCTION

Numerous problems in surface science deal with molecular motions [1-11] and relaxation phenomena [12-18] at fluid interfaces containing surface active substances (in short surfactants).

Under given conditions, tangential forces may exert in the interface of two liquids, together with the normal pressure. If the surface tension, σ , of the liquid interface changes from point to point, a tangential force will be exerted in addition to the pressure normal to the surface and its magnitude is determined by the surface tension gradient [1], which per unit area is $\vec{p}_t = \text{grad } \sigma$. The plus sign preceding the gradient indicates that this force tends to move the surface of the liquid in a direction from lower to higher surface tension.

There are many examples where the presence of a surfactant has an important role. Probably the best known is the effect of a surfactant on a liquid drop, immersed in a bulk liquid, initially at rest [2, 3]. The force acting on the unit volume of the drop (with density ρ') immersed in a bulk liquid (of

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density ρ), will be cancelled $(\rho-\rho') \vec{g} = 0$ either when the densities of the two liquids are equal ($\rho'=\rho$) or in the absence of gravity $\vec{g} = 0$ (zero gravity). Such a drop is called “free” and is motionless.

Recently, we have presented both experimental data and theoretical approach [7, 8] on the motion of non-deformed and deformable free drops in a continuous medium. However, to provide accurate description for the movement of a free drop, including the deformation, the translation as well as the breaking up of the drop, the present study explores the drop shapes using dimensionless analysis and a new dimensionless number, which is related to the Reynolds number. Consequently, the Marangoni force in the dimensionless form acting on the drop is calculated.

HYDRODYNAMIC EQUATIONS

We shall consider a viscous liquid drop L' (density ρ') immersed in an immiscible bulk liquid L , (density ρ). If the two liquids have the same density, $\rho' = \rho$, the drop is called free and is motionless or at zero gravity. The two liquids outside and inside the drop (see Fig.1) are considered Newtonians, incompressible and viscous having the viscosities μ and μ' , respectively. The surface between the two liquids is characterized by an interfacial tension, noted σ_0 .

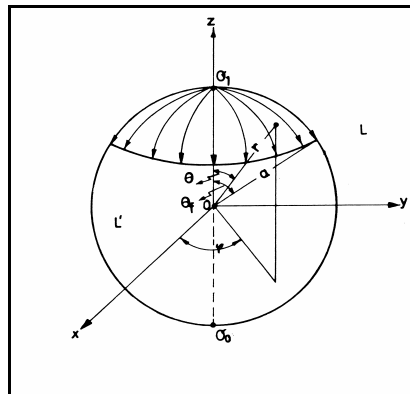


Figure 1. The spreading of a surfactant on a free drop surface

A small quantity of a surfactant (e.g., a droplet of 10^{-3} - 10^{-2} cm^3 , which is very small compared with the volume of the initial drop) is introduced in a point (called injection point) on the drop surface. The surfactant, because of its molecular structure, is spread and simultaneously adsorbed at the liquid-liquid interface and it is continuously swept along the meridians of the drop, by the convective transport. In the injection point the interfacial tension is

instantaneously lowered to σ_1 ($\sigma_1 < \sigma_0$) value. Since the interfacial tension is a function of the surfactant concentration, a gradient of interfacial tension is established over the surface of the drop. Consequently, the Marangoni spreading of the surfactant takes place from low surface tension to high surface tension.

The equations governing the flow considered quasi steady and axisymmetric [4] are the continuity equations for an incompressible fluid

$$\nabla \cdot \bar{v} = 0, \quad (1)$$

$$\nabla \cdot \bar{v}' = 0, \quad (2)$$

where \bar{v} and \bar{v}' are the velocities of the bulk liquid L, respectively, of the liquid L' within the drop, and the Navier- Stokes equations, for a steady flow

$$(\bar{v} \cdot \nabla) \bar{v} = -\frac{1}{\rho} \nabla p + \nu \Delta \bar{v}, \quad (3)$$

$$(\bar{v}' \cdot \nabla) \bar{v}' = -\frac{1}{\rho} \nabla p' + \nu' \Delta \bar{v}', \quad (4)$$

where p, p' are the pressures, outside and inside the drop and $\nu = \frac{\mu}{\rho}$,

$\nu' = \frac{\mu'}{\rho}$ are the kinematic viscosities of continuous liquid and drop liquid, respectively.

The equation of the interfacial flow [5, 6] is given by

$$\Gamma(\bar{w} \cdot \nabla_s) \bar{w} = \bar{F} + \nabla_s \sigma + (\kappa + \varepsilon) \nabla_s (\nabla_s \cdot \bar{w}), \quad (5)$$

where $\bar{w} = \bar{v}_s$ is the interface velocity, $\bar{F} = \Gamma \bar{g} + \bar{T} - \bar{T}'$ is the external force acting on the drop surface, \bar{T} and \bar{T}' are the tractions exerted by the outer and inner liquid on the drop interface, Γ is the surface density, κ and ε are the surface dilatational and shear viscosity, respectively, and ∇_s is the surface gradient operator. Because the surface density is very small ($\Gamma \approx 10^{-7} \text{g cm}^{-2}$) the inertial term in (5) can be neglected against the remainder terms.

In order to find the distributions of the velocities \bar{v}, \bar{v}' and of the pressures p, p' , the system of Eqs (1)-(5) must be solved taking into account some appropriate boundary conditions [4]. Some details are recently given by our group [7, 8].

The symmetry of the problem suggests a system of spherical coordinates (r, θ, φ) with the origin placed in the drop center and with the Oz axis passing through the sphere, (see fig. 1) in the point of the minimum interfacial tension, i. e., the injection point of the surfactant.

We underline that the surfactant injection point at the drop surface may be taken anywhere, the drop being initially at rest. In the following, we shall take it like shown in Fig. 1. The surfactant front position, in this radial flow, is noted by the angle θ_f .

The interfacial tension, σ , is considered a unique function of the angle θ . Within the surfactant invaded region, ($0 \leq \theta \leq \theta_f$), for the variation of the interfacial tension with θ , we take [9]

$$\sigma(\theta) = \frac{\sigma_0 - \sigma_1}{1 - \cos \theta_f} (1 - \cos \theta) + \sigma_1, \quad (6)$$

where $\sigma_0 = \sigma(\theta_f)$ and $\sigma_1 = \sigma(0)$.

Derivation of Eq. (6) gives the interfacial tension gradient in the invaded drop region with surfactant

$$\frac{d\sigma}{d\theta} = \frac{\sigma_0 - \sigma_1}{1 - \cos \theta_f} \sin \theta, \quad (7)$$

where, the interfacial tension, σ_0 , is constant in any point of the uncovered surface, while the interfacial tension difference, Π , also called surface pressure is

$$\Pi = \sigma_0 - \sigma_1, \quad (8a)$$

or

$$\Pi = \sigma_0 (1 - \beta), \quad (8b)$$

where β is the interfacial tensions ratio

$$\beta = \frac{\sigma_1}{\sigma_0}, \quad (9)$$

and arises only in the invaded region. It is clear that only σ_1 and σ_0 , i. e. the minimum and the maximum values of the interfacial tension, can be experimentally measured.

The surface flow leads to a stream of liquid directed to the drop along the Oz axis. This stream arises as a consequence of the continual replacement of that liquid layer which has been displaced by the surface flow, like a ventilation effect [3]. We immediately find that as the flow occurs with the driving by viscosity of the outer liquid L, forces of hydrodynamic pressure will act on the drop L'. The resultant of the forces exerted by the fluid on the drop, F_M , due to the symmetry of the Marangoni flow is oriented along the Oz axis and may be calculated from the general expression of force [4] by integration on the covered drop surface:

$$F_M = \iint_S [(p_{rr})_{r=a} \cos \theta - (p_{r\theta})_{r=a} \sin \theta] ds, \quad (10)$$

where S is the surface covered with surfactant, ds is the surface element, and p_{rr} , $p_{r\theta}$ are the normal and tangential components, of the viscous stress tensor [4] on the surface.

DIMENSIONLESS ANALYSIS

In dimensionless analysis, a dimensionless quantity (or more precisely, a quantity with the dimensions of 1) is a quantity without any physical units and thus is a pure number. Such a number is typically defined as a product or ratio of quantities which do have units, in such a way that all the units cancel out. For the dimensionless analysis of a mathematical model of a physicochemical phenomenon, the equations that describe the phenomena must be expressed in dimensionless form and therefore all dimensional variables that appear in the hydrodynamics equations must be expressed in terms of characteristic factors of these variables, named the scaling process. In our case the radius (a) of the drop is a characteristic length.

All linear dimensions can therefore be dimensionless ratios, of the form $\bar{r} = \frac{r}{a}$. In the same manner, the initial interface tension σ_0 may be considered as the characteristic dimension of the interfacial tension so that we have the dimensionless surface tension $\bar{\sigma} = \frac{\sigma}{\sigma_0}$.

Because the drop is initially at rest, we don't possess a characteristic velocity, U_c , so that we shall introduce one, expressed with the aid of some characteristic data of our problem. With these scale references we have the following dimensionless variables:

$$\bar{r} = \frac{r}{a}, \quad \bar{v} = \frac{v}{U_c}, \quad \bar{p} = \frac{p}{\rho U_c^2}, \quad \bar{\sigma} = \frac{\sigma}{\sigma_0}, \quad \bar{F} = \frac{F}{\rho a^2 U_c^2}, \quad (11)$$

where \bar{v} is the dimensionless velocity, \bar{p} represents the dimensionless pressure, $\bar{\sigma}$ is the dimensionless interface tension and \bar{F} stands for the dimensionless force.

The substitute of the dimensionless variables (11) in equations (1, 2) gives

$$\nabla \cdot \bar{\vec{v}} = 0 \quad (12)$$

$$\nabla \cdot \bar{\vec{\tau}} = 0 \quad (13)$$

the dimensionless continuity equations. In the same manner from (3, 4) we have

$$(\bar{\vec{v}} \cdot \nabla) \bar{\vec{v}} = -\nabla \bar{p} + \frac{1}{Re} \Delta \bar{\vec{v}} \quad (14)$$

$$(\bar{\vec{v}}' \cdot \nabla) \bar{\vec{v}}' = -\nabla \bar{p}' + \frac{1}{Re'} \Delta \bar{\vec{v}}' \quad (15)$$

for the Navier-Stokes dimensionless equations. Here, $Re = \frac{\rho a U_c}{\mu}$ and

$Re' = \frac{\rho a U_c}{\mu'}$ are the Reynolds numbers for the outside and inside flows.

If we consider the outer flows with Reynolds number equal to unity, $Re = 1$ we have

$$U_c = \frac{\mu}{\rho a} \quad (16)$$

This characteristic velocity, U_c , is called sometimes the viscous velocity.

For the flow inside the drop we'll obtain for the Reynolds number

$$Re' = \frac{\mu}{\mu'}. \quad (17)$$

With the values of the viscosities taken from [3, 8], the Reynolds number corresponding to the drop phase ranges between 1/80 and 1/2; this means that the Reynolds number is less than unity $Re' < 1$. Introducing the ratio of the bulk viscosities [10], $\lambda = \mu'/\mu$, we have also

$$Re' = 1/\lambda. \quad (18)$$

The velocities of the inner and outer liquid of the drop must satisfy the following kinematic conditions:

- the outer velocity must be zero far from the drop surface,
 $\vec{v} = 0$ for $\bar{r} \rightarrow \infty$;
- the normal component of the outer and the inner velocities must be zero on the surface of the drop
 $\vec{v}_n = \vec{v}'_n = 0$, at $\bar{r} = 1$
- the tangential velocity components of the two liquids at the interface must be continuous:
 $\vec{v}_t = \vec{v}'_t$ at $\bar{r} = 1$
- the velocity \vec{v}' within the drop must remain finite at all points, particularly at the centre of the drop ($\bar{r} = 0$ the origin of the coordinates).
- In addition to these kinematic conditions, a dynamic condition must be fulfilled at the interface, given by (5) in dimensionless form.

Eqs. [12-15] with these appropriate boundary conditions lead to the distribution of the velocity \vec{v} and of the pressures \bar{p} , outside the drop:

$$\bar{v}_r = \frac{\text{Ch } A}{(1 - \cos \theta_f)} \left(\frac{1}{\bar{r}^3} - \frac{1}{\bar{r}} \right) \cos \theta, \quad (19)$$

$$\bar{v}_\theta = \frac{\text{Ch } A}{(1 - \cos \theta_f)} \left(\frac{1}{2\bar{r}^3} + \frac{1}{2\bar{r}} \right) \sin \theta, \quad (20)$$

$$\bar{p} = -\frac{\text{Ch } A}{(1 - \cos \theta_f)} \frac{1}{\bar{r}^2} \cos \theta, \quad (21)$$

where A is a dimensionless constant of the following form:

$$A = \frac{(1 - \beta)}{3(1 + \lambda)}. \quad (22)$$

Here, we have introduced a new dimensionless number, Ch, given by:

$$\text{Ch} = \frac{\sigma_0 \rho a}{\mu^2}. \quad (23)$$

We named this new dimensionless number, Ch, Chifu's number, as recognition of Chifu's excellent scientific contributions to the examination and characterization of the behavior of liquids under the action of surface tension gradients caused by various surfactants or by differences in temperature, under the normal conditions or in conditions of microgravity [19- 21].

We have to observe that for the proposed characteristic velocity, U_c , we have for the Ch number the following connections with well-known dimensionless numbers, such as capillary number, Ca, Ohnesorge number,

Oh, and Weber number, We. So, we have $\text{Ch} = \frac{1}{\text{Ca}}$ where, $\text{Ca} = \frac{\mu U_c}{\sigma_0}$ is

the capillary number, $\text{Ch} = \frac{1}{\text{Oh}^2}$, for $\text{Oh} = \frac{\mu}{\sqrt{\sigma_0 a \rho}}$ being the Ohnesorge

number and $\text{Ch} = \frac{1}{\text{We}}$, where $\text{We} = \frac{\rho U_c^2 a}{\sigma_0}$ is the Weber number.

Further, we mention here, that the surface tension σ usually depends on the scalar fields, applied to the system (e.g., the temperature field and the electrical field) as well as on the concentration of foreign materials on the surface (so named surfactants). In the present paper, we focus on the variation due to surfactants (foreign materials) given by $\sigma(\theta)$, which deepens not only on θ but also on σ_0 , σ_1 , μ and μ' , all being concentrate in constant A.

The Marangoni force acting on the drop in dimensionless form is

$$\bar{F}_M(\theta_f) = \frac{2\pi \text{Ch} (1 - \beta)}{3(1 + \lambda)} (1 - 2\cos \theta_f - 2\cos^2 \theta_f). \quad (24)$$

Eq. (24) represents the dimensionless Marangoni force acting on the drop surface along the Oz axis. It can be seen that this force depends on the θ_f angle, namely, on the extent to which the drop surface is covered by the surfactant, the interfacial tension ratio, β , the ratio of the viscosities, λ , and the dimensionless number Ch.

From (24) it is observed that the force acting on the drop depends direct proportionally on the number Ch.

For further discussions of the dimensionless Marangoni force, it is useful to introduce the function

$$f(\theta_f) = 1 - 2 \cos \theta_f - 2 \cos^2 \theta_f . \quad (25)$$

The value of θ_f , for which $f(\theta_f) = 0$, is noted by θ_0 , and its value is $\theta_0 \approx 68.53^\circ$. This θ_0 is the value of θ_f for which the resultant force cancels. Also, we note that for $\theta_f \in [0, \theta_0)$, this function is negative $f(\theta_f) < 0$, and for $\theta_f \in (\theta_0, 180^\circ]$ the function is positive $f(\theta_f) > 0$, having the greatest value for $\theta_m = 120^\circ$.

From Eq. (24) it is found that for a coverage degree $\theta_f < \theta_0$ of the drop with surfactant, as a result of the appearance of interfacial tension gradient, the pressure force \bar{F}_M exerted by the external liquid upon the drop is oriented toward the negative direction of the Oz axis (Fig. 1). This is similar with the application of a "hammer" knock on the drop in the injecting point of the surfactant. For a coverage degree θ_f greater than θ_0 , the force \bar{F}_M is oriented towards the positive direction of the Oz axis. This is the propulsive (lifting) resultant force $\bar{F}_M > 0$, responsible for the upward movement of the drop.

The action of the Marangoni force \bar{F}_M on the drop is maximum at the injection point of the surfactant, for which $\theta_f = 0$

$$\bar{F}_M(0) = \frac{2\pi Ch(1-\beta)}{1+\lambda}, \quad (26)$$

where for physical meanings, we have taken its absolute value. The Marangoni force, noted also $\bar{F}(0)$, produces the hammer effect, identified earlier by us [7, 8]. It depends on Ch number, viscosities ratio, λ , and surface tension ratio, β .

RESULTS AND DISCUSSION

In the following, we present the physical and chemical characteristics of the six investigated experimental systems (Table 1). Each system contains a free drop, initially at rest. Upon the surfactant injection in a particular point on the drop surface, we have experimentally observed the shape modifications and the movement of the drops in time [8].

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Table 1. Composition and physical characteristics of the liquid/liquid systems. The drop radius (a) is 1.19 cm for systems 1 and 2 and 0.46 cm for systems 3 to 6.

System No.	Density g/cm^3	Continuous Phase (L)			Drop Phase (L')		Surfactant Solution (S)	
		Composition (% vol)	μ (cP)	σ_0 (L/L') (dyn/cm)	Composition (% vol)	μ' (cP)	Composition (% vol)	σ_1 (L'/S) (dyn/cm)
1	0.863	Ethanol 78.6 Water 21.4	2.26	7.9	Paraffin oil	80	Propanol 77.3 Water 22.7	3.5
2	0.863	Methanol 78 Water 22	1.33	10.2	Paraffin oil	80	Propanol 77.3 Water 22.7	3.5
3	1.068	NaNO ₃ 15.1 Water 84.9	1.1	28.7	Chlor benzene 40 Silicon oil 60	8.04	Benzylic alcohol 89 CCl ₄ 11	3.6
4	1.068	NaNO ₃ 15.1 Water 84.9	1.1	28.2	Chlor benzene 50 Silicon oil 50	5.46	Benzylic alcohol 89 CCl ₄ 11	3.6
5	1.066	NaNO ₃ 15 Water 85	1.1	25.6	Chlor benzene 85 Silicon oil 15	1.40	Benzylic alcohol 89 CCl ₄ 11	3.5
6	1.064	NaNO ₃ 14.9 Water 85.1	1.1	22.8	Chlor benzene 92 Silicon oil 8	1.03	Benzylic alcohol 89 CCl ₄ 11	3.6

Further, we examine the shapes, positions and movements of the six freely suspended drops at zero density difference between the drop and the suspending (continuous) medium. Each free drop is under a different interfacial tension gradient acting on the drop surface. The drop dynamics is investigated as a function of the drop to medium viscosity ratio, λ , the surface tension ratio, β , and the surface pressure, Π .

Furthermore, the experimental data are explored using non-dimensional analysis, and consequently, the dimensionless values of Ch number and of Marangoni hammer force, $\bar{F}(0)$, are calculated and are also given in Table 2.

The surfactant film, spread and adsorbed on the drop surface, exerts a certain surface pressure, Π , which is given by the difference between the interfacial tension σ_0 , which is constant in any point of the uncovered surface, and the interfacial tension σ_1 at the injection point with surfactant at the drop surface (Fig. 1). The experimental observations are given in Table 2 for the six investigated systems.

Table 2. Surface pressure, Π , the viscosities ratio, λ , the ratio of surface tensions, β , non-dimensional number, Ch (Eq. (23)), and the Marangoni hammer force, $\bar{F}(0)$ (Eq. (26)), in its non-dimensional form, for the six chosen systems, given in Table 1.

System No.	Π (dyn/cm)	λ	β	$Ch \times 10^{-4}$	$\bar{F}(0) \times 10^{-4}$	Experimental Observations
1	4.4 ± 0.3	35.39	0.44	1.5884	0.1535	The drop remains practically undeformable and motionless. Experimental work is given in [8].
2	6.7 ± 0.3	60.15	0.34	5.9218	0.4014	
3	25.1 ± 0.3	7.31	0.12	11.6527	7.7494	The drop shows deformations but after 0.6-0.8 sec., it returns to its initial form. A translation motion is also observed [8].
4	24.6 ± 0.3	4.96	0.13	11.4497	10.4961	
5	22.1 ± 0.3	1.27	0.14	10.3746	24.6833	The drop, after 0.3-0.4 sec., breaks up into two droplets. The resulted droplets have translation motion [8].
6	19.2 ± 0.3	0.94	0.16	9.2225	25.0776	

Analyzing the data from Table 2, it is to be noticed that the low values of Marangoni hammer force corresponding to low Ch numbers will not have a strong effect on the drop (see, cases 1 and 2).

At medium values of both Ch and $\bar{F}(0)$, the drops are deformable but they will reach a steady state shape with a small translational movement (see, cases 3 and 4, given in Table 2).

At very high values of the Marangoni hammer force $\bar{F}(0)$ and still high Ch numbers the deformations and particularly the elongations of the drop are high resulting in the breaking up of the drop (cases 5 and 6, given in Table 2).

For chosen λ and Π values, and for an initial drop position, drops show increasingly pronounced deformations with increasing Ch number (cases 3 and 4, Table 2). For very high λ (cases 1 and 2, Table 2) the drops are nondeformable.

At λ approximately 1, for high Ch numbers, the drops show increasingly pronounced deformations. Then, a continued drop elongation brings the possible onset of drop break up. The last effect is observed for sufficiently large Ch numbers and Π values and for low λ values (cases 5 and 6, Table 2).

As shown above, the movements of free drops depend on Ch number, viscosities ratio, λ , the surface pressure, Π , and the Marangoni hammer force, $\bar{F}(0)$. From theory and experimental work it is clear that λ ratio has a critical role for drop deformations and translation movement.

Thus, by using the dimensionless analysis, the dimensionless values of Ch number and of Marangoni force $\bar{F}(0)$ were calculated and consequently, the deformations and the movement were accurately described for free drops, at low Reynolds numbers.

CONCLUSIONS

The movement of free drops, such as deformations, translation motion and break up of the free drops, was explored using dimensionless analysis, a new dimensionless number, Ch, which is related with Reynolds numbers, and the Marangoni hammer force, $\bar{F}(0)$ in its dimensionless form.

We found that the movement of free drops suspended in a continuous medium depends on the viscosity ratio, λ , the surface pressure, Π , and dimensionless values of Ch number and of the Marangoni hammer force, $\bar{F}(0)$.

For very viscous drops and not so high values of Π , Ch number and of $\bar{F}(0)$, the drops remain undeformable and a very small translational motion is observed (cases 1 and 2, given in Table 2).

At medium values of $\bar{F}(0)$ and λ , and high values of both surface pressure, Π , and Ch number, the drops are deformable but they will reach a steady state shape with a small translational movement (cases 3 and 4, given in Table 2).

At very high values of the Marangoni hammer force $\bar{F}(0)$ and still high values of Ch numbers and of surface pressure, Π , but for low values of λ , the deformations and particularly the elongations of the drops are high resulting in the braking up of the drops (cases 5 and 6, given in Table 2).

Certainly, the obtained results can be attributed to a complex mechanism including the surface dilution and tip-stretching of the surface tension, as well as capillary forces.

In our opinion, the real flow at the drop interface causes the motion of the neighbouring liquids by viscous traction and generates the Marangoni force, which is the principal factor that develops the deformation and the translational motion of the free drops.

The results of our theoretical hydrodynamic model are in a substantial agreement with the observed experimental data.

EXPERIMENTAL SECTION

The experimental work on the drop dynamics was performed in liquid-liquid systems of equal densities and recently published by us [8].

The mixtures, making up the continuous L phase, were placed in a thermostated vessel of 1 dm³, made of transparent glass. The drop (L') was made of various radii between 0.46 and 1.19 cm, by using the mixtures described in Table 1.

After the system was stabilized, a small quantity (10⁻³-10⁻² cm³) of the surfactant solution (S) was injected with a micrometric syringe, in a point on the drop surface (injection point in Fig. 1).

The interfacial tension for the liquid/liquid systems, e.g. L/L' and L'/S, was determined by a method based on capillarity [3] and its value is given in Table 1 at constant temperature (20 ± 0.1°C) as described in [8]. The position of the surfactant front and the sequences of the drop dynamics for the six investigated systems are also presented elsewhere [8].

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