Recommendations on determination of interfacial tension at the interface between two fluids by the spinning drop method

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Abstract

There are studied popular methods of B. Vonnegut, H. Princen, J. Slattery, S. Torza for measuring the interfacial tension at the interface between two immiscible fluids by the spinning drop method. There is provided the method for calculating the geometric parameters of the spinning drop, and its results to the emergence of a spinning drop at the centre of the severely cylindrical area that correspond to the range of the ratio of the cube of the drop`s length to its volume of 24–120, and the range of the ratio of the spinning drop`s length to its diameter 4.00–0.35.

Based on the obtained calculation results there is offered the method for determining the interfacial tension by using approximation dependence of the interfacial tension from the given drop volume, its length, difference in fluid densities and the angular velocity of the drop rotation. There are assessed methodological errors of the offered and the known methods for determining interfacial tension according to the rotating drop method. A flow chart and the overall appearance of the device that implements the proposed method for measuring the interfacial tension is presented.

Keywords: *a spinning drop, interfacial tension, length of the drop, measurement, methodological error, the device, volume of a drop.*

Introduction

During some processes, such as extraction of oil, an interfacial tension (IT) at the interface of water or aqueous surfactant and reservoir oil is an important physical and chemical parameter [1]. It should be noted that the value of the IT at the specified interface can be in the range of 0.01 and 20 mN/m. Low values of the IT (0.01 or less) can be measured only by devices that implement the spinning drop method [2].

The spinning drop method means that the horizontally placed glass tube is filled with the heavier fluid under consideration, such as aqueous surfactant, and a drop of the lighter fluid under consideration, such as oil, is injected into this fluid, and then the tube is turned around its horizontal axis with a certain angular velocity ω. The appropriate dimensions of the spinning drop are measured depending on the method for determining interfacial tension (e.g. its largest diameter, length, volume) and the difference of contacting fluids densities $\Delta\rho$, and the value of interfacial tension σ is calculated based on the appropriate dependencies [3–6].

Now the widespread dependencies, regardless of the date of their publication, are as follows:

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B. Vonnegut dependency [3]

$$\sigma = \Delta\rho\omega^2 R^3 / 4, \qquad (1)$$

H. Princen dependency [4]

$$\frac{x_0}{r} = \frac{2}{3}\frac{cr^3+1}{\left(cr^3\right)^{1/3}}, \qquad (2)$$

J. Slattery dependency [5]

$$\sigma = \left(R/r^*\right)^3 \Delta\rho\omega^2 / 2, \qquad (3)$$

S. Torza dependency [6]

$$\sigma = \pi^{-3/2}\Delta\rho\omega^2 \left(V/x_0\right)^{3/2} / 4, \qquad (4)$$

where $\Delta\rho = \rho_1 - \rho_2$ is the densities difference of heavy and light liquids, respectively; R is the largest real radius of the spinning drop; x_0 is the half of the length of the spinning drop; $r = \sqrt[3]{3V/(4\pi)}$ is the radius of the sphere of lighter fluid of the volume V, which is injected in a tube with the heavier fluid; $c = \Delta\rho\omega^2/(4\sigma)$ is a characteristic parameter, which is the basis for calculating IT σ according to H. Princen method; r^* is a dimensionless parameter, which is determined based on J.Slattery Table [5], depending on the ratio R/x_0 (Table 1).

It should be noted that B.Vonnegut suggests to apply the dependence (1) on the condition that $x_0/R > 4$ [3]. H. Princen suggests to apply the dependence (2) on the condition that $x_0/R > 3.645$ [4]. According to J.Slattery [5] if $x_0/R > 4$, the dependence

Table 1 – The data of the dependence $r^* = f\left(R/x_0\right)$ [5]

R/x_0	r^*	R/x_0	r^*	R/x_0	r^*
1.0000	0	0.7415	1.0000	0.2708	1.2570
0.9997	0.1	0.6432	1.1000	1.2543	1.2580
0.9980	0.2	0.4928	1.2000	0.2297	12590
0.9932	0.3	0.3332	1.2500	0.2262	1.2591
0.9840	0.4	0.3268	1.2510	0.2225	1.2592
0.9687	0.5	0.3198	1.2520	0.2183	1.2593
0.9459	0.6	0.3122	1.2530	0.2136	1.2594
0.9140	0.7	0.3038	1.2540	0.2081	1.2595
0.8710	0.8	0.2945	1.2550	0.2016	1.2596
0.8148	0.9	0.2837	1.2560	0.1932	1.2597

(3) has a methodological error that is less than 0.4%. S. Torza suggests to apply the dependence (4) for $cr^3 > 100$ [6], which corresponds to the ratio $x_0 / R > 67$.

Therefore it is necessary to assess methodological errors of the mentioned methods for calculating IT σ by the spinning drop method, to develop recommendations for their application.

Theoretical part

To assess the methodological errors of the above mentioned methods we have made theoretical calculations of geometrical dimensions of the spinning drop.

Let us consider a horizontal spinning tube 1, filled with the fluid 2 with greater fluid density, and the fluid drop 3 with a lower density ρ_1 (Fig. 1). We assume that the pressure at the axis y inside the drop (O) equals p_{O1}, and outside the drop – p_{O2}. In this case, the force of gravitaty is neglected, and it is assumed that the axes of rotation of the tube 1 and the drop 3 coincide.

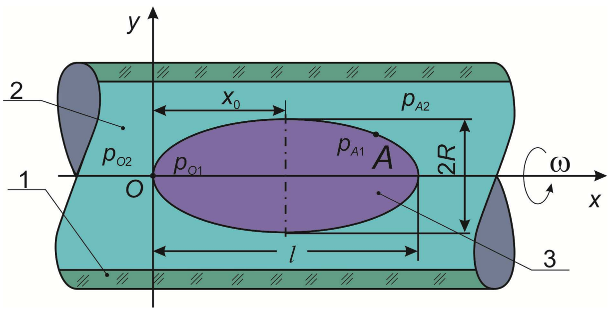

Figure 1 – The rotary tube filled with the heavier and lighter fluids under consideration

Therefore the pressure p_{A1} at point A in the middle of the drop is the following:

$$p_{A1} = p_{O1} + \rho_1 y^2 \omega^2 / 2, \tag{5}$$

where y is the distance from the point A to the axis x.

Similarly, the pressure outside the drop at the point A is as follows:

$$p_{A2} = p_{O2} + \rho_2 y^2 \omega^2 / 2. \tag{6}$$

Therefore pressures difference at the boundary of two fluids at the point A is as follows:

$$p_{A1} - p_{A2} = p_{O1} - p_{O2} - \Delta\rho y^2 \omega^2 / 2. \tag{7}$$

Due to the gravitational force the rotation axis of the drop shifts to the rotation axis of the tube at the value of $y^* \approx R^2 \Delta\rho g / (\eta\omega)$ [6], where g is the acceleration of gravity, η is the dynamic viscosity of the heavier liquid. But the shape of the spinning drop does not change in this case.

On the other side, the pressures drop across the phases interface at the point A is as follows:

$$p_{A1} - p_{A2} = \sigma\left(1/R_1 + 1/R_2\right), \tag{8}$$

where R_1 i R_2 are the radii of curvature of the drop surface at the point A of the plane of Fig. 1 and at the plane perpendicular to the plane of Fig. 1, respectively [7].

Besides, the pressures difference Δp_0 across the phases interface at the level of the horizontal rotational axis x at the point O is as follows [7]:

$$p_{O1} - p_{O2} = \Delta p_0 = 2\sigma / R_0, \tag{9}$$

where R_0 is the radii of curvature of the interface of the spinning drop at the point O (Fig. 1).

Therefore having regard to the (8) and (9) the dependence (7) is as follows:

$$\sigma\left(1/R_1 + 1/R_2\right) = 2\sigma / R_0 - \Delta\rho y^2 \omega^2 / 2. \tag{10}$$

The equation (10) is a strict equation that describes the shape of the spinning drop depending on σ, $\Delta\rho$ and ω in the absence of gravitational force.

If $R_1 = ds / d\varphi$, $R_2 = y / \sin\varphi$, where s is the length of the profile arc of the spinning drop, φ is the angle between the axis x and the normal to the point A at the surface area of the spinning drop [7], then the equation (10) is as follows:

$$\frac{d\varphi}{ds} = \frac{2}{R_0} - \frac{\omega^2 y^2 \Delta\rho}{2\sigma} - \frac{\sin\varphi}{y}. \tag{11}$$

Having introduced a new variable $a^3 = \sigma / \left(\Delta\rho\omega^2\right) = 1/\left(4c\right)$, we obtain

$$\frac{d\varphi}{ds} = \frac{2}{R_0} - \frac{y^2}{2a^3} - \frac{\sin\varphi}{y}. \tag{12}$$

Having multiplied the left and right parts (12) by *a*, we obtain the equation in a dimensionless form that describes the surface area of the spinning drop:

$$\frac{d\varphi}{d(s/a)} = \frac{2}{R_0/a} - \frac{1}{2}\left(\frac{y}{a}\right)^2 - \frac{\sin\varphi}{y/a}. \qquad (13)$$

Other variables, included in (13), can be calculated using the following dependencies [7]:

$$\frac{d(y/a)}{d(s/a)} = \cos\varphi,$$

$$\frac{d(V/a^3)}{d(s/a)} = 2\pi\left(\frac{y}{a}\right)^2 \sin\varphi, \qquad (14)$$

$$\frac{d(x/a)}{d(s/a)} = \sin\varphi.$$

Having solved the dependencies (13) and (14) for different values of R_0/a at the moment of reaching the angle $\varphi = 90^\circ$, we calculate the appropriate geometrical parameters of the spinning drop.

The initial conditions in this case are as follows:

$$y = x = s = V = \varphi = 0,\ 1/R_0 = 1/R_1 = 1/R_2; \qquad (15)$$

and the final ones are:

$$R/a = 4^{1/3},\ R_0/a = 2\cdot 4^{1/3}/3,\ R/R_0 = 3/2. \qquad (16)$$

If the final conditions (16) are reached, the parameters do not increase according to (16), and the surface area of the spinning drop is strictly cylindrical in its central part, i.e. $R_1 \to \infty$, $R_2 = R$.

The results of calculation and their discussion

The results of calculation of the dimensionless parameters of a spinning drop (a^3/V, l^3/V, R/r, cr^3, $l/(2r)$ by the Runge – Kutta method of the 4th order equations (13) and (14), taking into account (15) and (16) for $1.05826519 \le R_0/a \le 1.05826736797879$ and $\varphi = 90°$ that correspond to the relation $l/(2r) \ge 4$, are shown in Table 2, where $l = 2x_0$. It should be noted that the calculation was carried out with an error of the end values $2.22\cdot 10^{-16}$. Besides, during the process of calculation there is preserved the 32 width of geometrical values of the spinning drop parameters.

The calculation results a^3/V for the range of $l^3/V = 24-120$, which corresponds to the variable $l/(2R)$ in the range of $4.00-9.35$, were applied for obtaining the approximation dependence $a^3/V = f(l^3/V)$ of the following type:

$$\frac{a^3}{V} = \frac{A(l^3/V)^2 + B(l^3/V) + C}{l^3/V + D}, \qquad (17)$$

where $A = -5.893\cdot 10^{-6}$, $B = 0.003261$, $C = 0.259$, $D = 3.648$.

Then the value σ of the IT can be calculated in the following way:

$$\sigma = \Delta\rho\omega^2 V\left(\frac{A(l^3/V)^2 + B(l^3/V) + C}{l^3/V + D}\right). \qquad (18)$$

It should be noted that in this case the spinning drop in the central part is not strictly cylindrical and has the form of an elongated ellipse. It is offered to carry out the measurement of the interfacial tension σ by the spinning drop method at low angular velocities of the drop`s spinning that result in emergence of the strictly cylindrical area at the drop`s central part that does not result in the device vibrations, which occur at drop spinning at high velocities.

The assessment of the relative methodological errors δ_{meth} of the offered approximation dependence (17) for all values $(l^3/V)_{\text{tab}}$ in a specified range of values was carried out in the following way:

$$\delta_{meth} = \left(\left(\frac{a^3}{V}\right)_{calc} - \left(\frac{a^3}{V}\right)_{tab}\right) \Big/ \left(\frac{a^3}{V}\right)_{tab}, \qquad (19)$$

where $(a^3/V)_{calc}$ and $(a^3/V)_{tab}$ are calculated according to the dependence (17) and the values in the Table (a^3/V) are correspondingly calculated for every value l^3/V under the Table 2. The mentioned relative methodological errors are shown in Table 2 and they do not exceed the absolute value of 0.00082, which satisfies the requirements for determining the accuracy of the IT by the spinning drop method.

It should be noted that in order to implement B. Vonnegut and J. Slattery methods it is necessary to measure the largest radius R of the spinning drop, which is significantly affected by the coefficient of the optical zoom M of the tube with the tested fluids:

$$M = R_{meas}/R, \qquad (20)$$

where R_{meas} is the largest measured radius of the spinning drop.

This coefficient can vary in the range of 1.332 to 1.340 [8]. Determining the specific value M depends on many factors and can lead to significant additional errors of the obtained results.

H. Princen dependence (2) provides that there was formed a strictly cylindrical area at the centre of the spinning drop during its rotation. The area appears at high angular velocities of the drop rotation.

The author [5] indicates that S. Torza dependence (4) is recommended to be used for the ratios $x_0/\mathrm{R} > 67$, which also require high angular velocities of the drop rotation. This leads to vibration of devices that implement the above mentioned H. Princen and S. Torza methods.

Therefore, during measuring the IT σ by the spinning drop method we recommend to achieve such angular velocities of the drop rotation, at which the ratio of the cube of its length l to the volume of the drop V is within the range of 24–120, corresponding to the ratio of the spinning drop length to its diameter 4.00–9.35.

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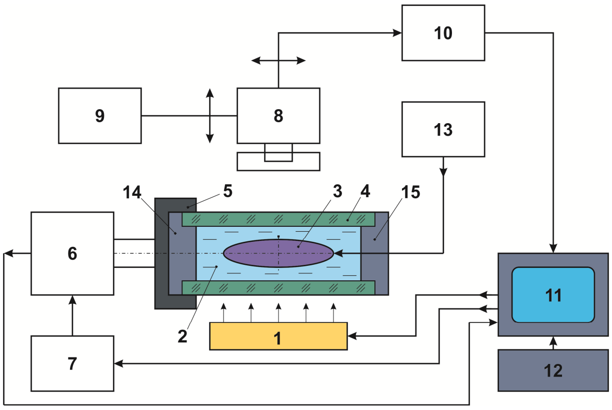
Table 2 – The calculation results of geometrical parameters of the spinning drop

R_0 / a	a^3 / V	$l / 2R$	l^3 / V	R / r	cr^3	$l / (2r)$	Relative error $\delta_{meth} \cdot 10^5$
1.05826519000000	0.012075	3.967485	24.027738	0.586213	4.942706	4.651580	81.00
1.05826544000000	0.011947	4.002410	24.413265	0.584189	4.995526	4.676327	63.00
1.05826569000000	0.011805	4.042163	24.855768	0.581918	5.055622	4.704411	54.00
1.05826594000000	0.011644	4.088375	25.375108	0.579319	5.125450	4.736950	31.00
1.05826619000000	0.011458	4.143560	26.002221	0.576274	5.208792	4.775656	8.30
1.05826644000000	0.011235	4.211987	26.790343	0.572583	5.312071	4.823426	14.50
1.05826669000000	0.010955	4.302137	27.846485	0.567857	5.448046	4.885995	36.50
1.05826694000000	0.010569	4.434309	29.431645	0.561192	5.647237	4.977001	56.40
1.05826719000000	0.009902	4.686718	32.580530	0.549267	6.027195	5.148518	70.40
1.05826736707000	0.007180	6.208348	54.975953	0.493645	8.312249	6.129435	62.10
1.05826736732000	0.007062	6.301242	56.533652	0.490918	8.451627	6.186788	54.00
1.05826736757000	0.006893	6.438921	58.882718	0.486985	8.658191	6.271318	69.00
1.05826736782000	0.006582	6.711838	63.681884	0.479546	9.067634	6.437267	82.00
1.05826736797009	0.005781	7.549940	79.605536	0.459237	10.324878	6.934428	92.00
1.05826736797034	0.005774	7.558256	79.772487	0.459052	10.337351	6.939272	1.60
1.05826736797059	0.005766	7.566886	79.945949	0.458861	10.350297	6.944298	0.71
1.05826736797084	0.005759	7.575957	80.128478	0.458660	10.363904	6.949579	0.25
1.05826736797109	0.005751	7.585154	80.313757	0.458457	10.377700	6.954931	1.25
1.05826736797134	0.005743	7.594603	80.504336	0.458248	10.391874	6.960428	2.25
1.05826736797159	0.005735	7.604241	80.698961	0.458036	10.406332	6.966033	3.28
1.05826736797184	0.005727	7.614698	80.910391	0.457806	10.422018	6.972111	4.33
1.05826736797209	0.005718	7.625155	81.122099	0.457577	10.437704	6.978187	5.46
1.05826736797234	0.005709	7.636368	81.349422	0.457331	10.454524	6.984699	6.58
1.05826736797259	0.005700	7.647518	81.575785	0.457088	10.471249	6.991172	7.77
1.05826736797284	0.005690	7.659487	81.819126	0.456827	10.489203	6.998116	8.94
1.05826736797309	0.005680	7.671582	82.065400	0.456564	10.507346	7.005131	10.20
1.05826736797334	0.005670	7.684685	82.332617	0.456279	10.527001	7.012726	11.40
1.05826736797359	0.005659	7.698102	82.606711	0.455989	10.547128	7.020499	12.80
1.05826736797384	0.005647	7.712339	82.898036	0.455682	10.568484	7.028742	14.10
1.05826736797409	0.005635	7.727584	83.210558	0.455353	10.591351	7.037564	15.60
1.05826736797434	0.005623	7.743269	83.532738	0.455017	10.614880	7.046635	17.10
1.05826736797459	0.005610	7.759774	83.872417	0.454664	10.639638	7.056174	18.60
1.05826736797484	0.005596	7.777223	84.232297	0.454291	10.665813	7.066251	20.20
1.05826736797509	0.005581	7.796059	84.621631	0.453891	10.694066	7.077122	21.80
1.05826736797534	0.005565	7.816406	85.043231	0.453460	10.724588	7.088855	23.50
21.05826736797559	0.005548	7.838202	85.496024	0.453000	10.757283	7.101414	25.30
1.05826736797584	0.005530	7.861321	85.977621	0.452514	10.791962	7.114723	27.20
1.05826736797609	0.005511	7.887149	86.517256	0.451974	10.830705	7.129577	29.10
1.05826736797634	0.005489	7.914867	87.098268	0.451397	10.872282	7.145501	31.20
1.05826736797659	0.005466	7.945797	87.748936	0.450757	10.918679	7.163251	33.40
1.05826736797684	0.005440	7.980886	88.490018	0.450035	10.971312	7.183360	35.60
1.05826736797709	0.005411	8.020131	89.322630	0.449233	11.030182	7.205819	38.00
1.05826736797734	0.005378	8.065740	90.295156	0.448308	11.098596	7.231877	40.40
1.05826736797759	0.005339	8.119349	91.445053	0.447231	11.179010	7.262446	42.90
1.05826736797784	0.005291	8.186754	92.901264	0.445891	11.280119	7.300794	45.30
1.05826736797809	0.005231	8.273372	94.789562	0.444192	11.410048	7.349927	47.50
1.05826736797834	0.005147	8.397851	97.536697	0.441795	11.596768	7.420256	49.00
1.05826736797854	0.005039	8.563025	101.242891	0.438693	11.844532	7.513075	48.10
1.05826736797874	0.004775	8.999145	111.362630	0.430902	12.498714	7.755492	40.40
1.05826736797875	0.004742	9.056786	112.736394	0.429913	12.585176	7.787252	22.00
1.05826736797876	0.004704	9.125893	114.394568	0.428739	12.688836	7.825246	36.00
1.05826736797877	0.004652	9.219631	116.663204	0.427167	12.829444	7.876637	54.00
1.05826736797878	0.004577	9.359544	120.091031	0.424862	13.039315	7.953038	82.00

Block diagram of the device that implements our methodology is shown in Fig. 2 and thus it allows measuring the IT by the spinning drop method.



1 – monochrome light source; 2 – heavier fluid under consideration; 3 – a spinning drop of the lighter fluid under consideration; 4 – a glass tube; 5 – collet holder; 6 – electronic engine; 7 – a unit of the length change of the spinning drop; 8 – measuring electron microscope; 9 – displace node; 10 – video input device; 11 – electronic computer unit; 12 – keyboard; 13 – a unit for the drop formation of a given volume; 14, 15 – plugs

Figure 2 – Block diagram of the device for measuring IT using the spinning drop method

Firstly, the glass tube 4, closed with the plug 14, is filled with the heavier liquid 2, for example, aqueous surfactant, and then the tube is filled with a drop 3 of the lighter liquid under consideration of the strictly specified volume V using the drop forming unit 13 of the lighter liquid under consideration, such as oil. A dosing microsyringe of the appropriate volume can be used as the unit 13. The glass tube is closed with the plug 15, preventing the formation of air bubbles in it, and it is horizontally set in a collet holder 5.

Then, using the keyboard 12, the electronic computer unit 11 is turned on and the process of measurement begins. As a result of this a constant voltage influences a unit of the length change of the spinning drop 7 causing the rotation of the output shaft of the motor 6 and the glass tube 4 filled with the tested fluids with a certain angular velocity ω. At the same time the monochrome light source 1 is turned on, resulting in passing of light rays through the glass tube 4 with the fluids and a video signal with the information about the form of the spinning drop 3 comes to the input of the measuring electron microscope 8. In order to measure the total length l of the spinning drop 3 a research operator manually sets the position of the measuring electron microscope 8 by the displace node 9 at the level, at which the image of the drop is the most contrast on the screen of the electronic computer unit 11, such as a computer, which passes through the video input device 10, and at this point it gives a signal through the keyboard 12 to measure the length l of the spinning drop 3.

Electronic computer unit 11 calculates the ratio l^3/V and controls the calculation of this ratio in the range of $24-120$ based on the information about the volume V of the drop and the density difference $\Delta\rho$ of the tested fluids, which are injected by the operator through the keyboard beforehand, and the input information about the angular velocity of the drop rotation ω and it`s length l. If the ratio is less than 24, then the voltage at the input of the block 7 automatically increases, resulting in the increase of angular velocity of the drop rotation and thus to the increase of the drop length l. If the ratio is greater than 120, then the voltage at the input of the block 7 decreases, resulting in reduction of ω and l, respectively. It should be noted that the electronic computer unit 11 informs the operator about the value of the ratio l^3/V.

If the ratio l^3/V is in the range of $24-120$, then the calculation of the IT σ is made based on the approximation dependence (8). It should be noted that the program provides for making measurements of IT σ for different values of the ratio l^3/V in the range of $24-120$. Additionally, the operator launches the measurement mode.

Information about the parameters l^3/V, ω, σ, $\Delta\rho$, V, l is shown on the monitor of the electronic computer unit 11 after the measurements are made, and a relevant document with the measurement results is formed in a database.

Fig. 3 shows the overall appearance of the device that implements the offered measurement method of IT σ.

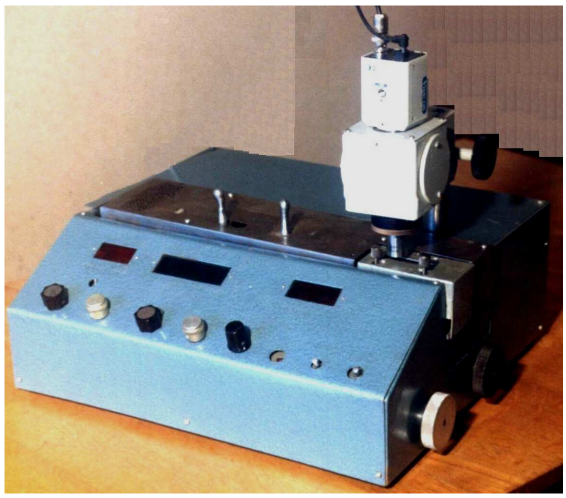


Figure 3 – The general view of the device for the measurement of the interfacial tension by the spinning drop method

It should be noted that the suggested measurement technique of IT σ and the device, which implements it, allow us to make measurements at low angular velocities of the drop rotation up to the formation of a strictly cylindrical section at its central part and they can also be used for obtaining the dependence of interfacial tension at the interface between two fluids at the time, which is important during the study and application of surfactant solutions, such as in technological processes of oil and gas production.

Conclusions

The spinning drop method is recommended to use for measuring the interfacial tension at the interface between two immiscible fluids when the values of interfacial tension are in the range of 0.01–20 mN/m or less. B.Vonnegut and J.Slattery methods for measuring interfacial tension by the spinning drop method provide that the ratio of the spinning droplet`s length to its diameter should exceed four, and a preliminary value of the coefficient of the optical zoom M of the glass tube with the tested fluids can range 1.332–1.340 and it has a significant influence on the methodological error of these methods. According to H.Prinsen and S.Torza methods the angular velocities of the drop rotation should have such values that there was formed a strictly cylindrical area in the central area of the drop. This requires substantial angular velocities of the drop rotation and leads to vibration of devices that implement the following methods.

The authors` method involves the application of the approximation dependence of the interfacial tension upon the given drop`s volume, densities difference of contacting fluids and the angular velocity of its rotation, which should not result in the formation of a strictly cylindrical area in the central part of the spinning drop, and the ratio of the cube of the spinning drop length to its volume should be in the range of 24–120, i.e. the device operates without vibration at low angular velocities of the drop rotation.

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Рекомендації щодо визначення міжфазного натягу на межі поділу між двома рідинами методом обертової краплі

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Розглянуті відомі методики Б.Воннегута, Г.Прінсена, Д.Слеттері, С.Торза вимірювання міжфазного натягу на межі розділу двох нерозчинних одна в одній рідин за методом обертової краплі. Наведено методику розрахунку геометричних параметрів обертової краплі, а також її результати до виникнення у центральній частині обертової краплі строго циліндричної ділянки, що відповідають діапазонам відношень кубу довжини краплі до її об'єму 24–120 і довжини обертової краплі до її діаметра 4.00–0.35.

На основі отриманих результатів розрахунку запропоновано методику визначення міжфазного натягу з використанням апроксимаційної залежності міжфазного натягу від заданого об'єму краплі, її довжини, різниці густин досліджуваних рідин і кутової швидкості обертання краплі. Оцінено методичні похибки запропонованої і відомих методик визначення міжфазного натягу. Наведено структурну схему і загальний вигляд приладу, який реалізує запропоновану методику вимірювання міжфазного натягу.

Ключові слова: *вимірювання, довжина краплі, методична похибка, міжфазний натяг, обертова крапля, об’єм краплі, прилад.*