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Motion of near-spherical micro-capsule in planar external flow

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Dynamics of a micro-capsule with compressible membrane placed into planar flow is considered. The form of the capsule is assumed to be near-spherical and the membrane forces are calculated in the first order in respect to the membrane displacement. We have established that the capsule dynamics is governed by two dimensionless parameters in this limit, which account for membrane stretch nodulus B, viscosities if inner fluid and solvent, capsule radius, external flow gradient and Taylor deformation parameter at rest. Phase diagram for capsule dynamical regimes is plotted on the plane of these two dimensionless parameters in the limits of low and high viscosity contrast between the fluid inside the capsule and the solvent.

1. Microcapsule is a soft particle closed with elastic shell filled with a fluid. The size of experimentally prepared capsules can reach 100 μm [1, 2] or millimeters [3]. Capsules represent a model of biological objects, most notably red blood cells, which explains a considerable scientific interest to the motion of capsules in external flows. It is also possible to use capsules as containers to deliver chemicals, mostly drugs, to a particular place in living organism [4]. At smaller scales this mechanism is realized in transport inside living cells with the help of membrane vesicles [5].

There are two types of objects which are described as capsules in scientific literature (see e.g. [6, 7]). The difference is in the properties of elastic membrane of capsules. In the present paper we theoretically examine the motion of capsules in an external flow assuming the shear and area compression modules of the membrane are of the same order. In such case membrane compressibility should be taken into account. This type of the capsules was experimentally investigated in [1–3]. The opposite limit of incompressible membrane with finite shear modulus is theoretically considered in [6, 8].

The experimental study of motion of nearly spherical capsules with compressible membrane in an external flow of solvent liquid was performed in [1–3]. It was reported that the capsule can experience tank-treading, which is accompanied by oscillations in inclination angle and shape (thus, the regime can be also called trembling). Mechanical properties of capsules and their motion in a flow determine rheological properties of a capsule suspension. For suspension of vesicles this connection was theoretically investigated in [9, 10] and the rheology of the suspension was measured in [11]. The mechanical properties of the capsule can be also of importance in the process of transport with the help the capsule since the capsule membrane should be broken only under particular conditions which ensure the delivery of substance to a destination.

The theoretical study of capsules had begun with the works [12, 13], and was continued by D. Barthès–Biesel in her following papers (see in particular [14, 15]). Numerical simulations for capsule in shear flow [7, 16] showed that capsule which have non-spherical form at rest can also undergo tumbling motion, if the shear strength and the inverse viscosity contrast λ between the solvent and the fluid filling the capsule and the solvent are not very large. Phase diagram was plotted for the capsule dynamical regimes on the plane of two dimensionless parameters, capillary number Ca and the viscosity contrast λ [7, 16]. Recently it was suggested phenomenological theory [17] and simplified numerical method [14] to explain the results for the capsules. All suggested schemes do not consider the general case of the planar external flow when the ratio between strain and rotational part of the flow can be arbitrary. The general case was studied for vesicles, both theoretically [18] and experimentally [19], and it was showed that there exist two dimensionless parameters which include also rotational part of the flow and Taylor deformation parameter of vesicle at rest.

In this letter, we develop systematic theory describing motion of capsules which have non-spherical shape at rest. We find the proper dimensionless parameters for capsules and plot the phase diagram. We consider influence of capsule non-sphericity at rest and corrections to angular velocity of the capsule. To our knowledge these two question were not theoretically studied before. Our results helps us to explain some of the experimental data reported in [1–3].

2. On the scales of the size of a capsule the Reynolds number for a fluid flow is very small. This allow us to omit both nonlinear and time derivative terms in Navier–Stokes equation. Thus the velocity field $\mathbf{v}$ is described by linear and quasi-stationary Stokes equation $\eta\Delta\mathbf{v} = \nabla P$, where P is pressure. We also consider a fluid to be incompressible: $(\nabla\cdot\mathbf{v}) = 0$.

In order to formulate the boundary conditions on the capsule shell one needs to consider the elastic forces acting on the membrane. Let us choose a Lagrange reference system ξ^α (where $\alpha = 1,2$) on the surface of the membrane. The current form of membrane is determined by the radius vector of an element: $r^i(\xi^\alpha)$, where index i is three-dimensional. We denote as $r_0^i(\xi^\alpha)$ the radius vector of the element in the unperturbed state. Since the capsule is capable of rotation as a whole it is essential for the theory of small deformations to introduce a rotation matrix $\mathcal{O}^{ij}$, so that:

$$r^i = R\mathcal{O}^{ij}(r_0^j + u^j), \qquad (1)$$

where $u^i(\xi^\alpha)$ is displacement vector of an element. Evolution of the rotation matrix is $\partial_t\mathcal{O}^{ij} = \Omega^{ik}\mathcal{O}^{kj}$. Angular velocity Ω^i ($\Omega^{ik} = \epsilon^{ikl}\Omega^l$, where ϵ^{ikl} is Levi–Civita symbol) of the capsule as a whole should be determined through equation $I^{ij}\Omega^j = M^i$ using found velocity field on the membrane surface, where I^{ij} is the inertia tensor of the membrane and M^i is its angular momentum assuming that the mass surface density is constant over all surface of the membrane at rest. We have also introduced a mean radius of a capsule so that $4\pi R^3/3$ is its volume which remains constant due to the impermeability of the membrane for the liquid. Both r_0^i and u^j are thus dimensionless.

The strain tensor in the linear approximation is:

$$u_{\alpha\beta} = \frac{1}{2}\left(\partial_\alpha r_0^i\,\partial_\beta u^i \;+\; \partial_\beta r_0^i\,\partial_\alpha u^i\right). \qquad (2)$$

In the same approximation the energy of deformations can be written as:

$$\mathcal{F} = \frac{1}{2}\int dA\left[Ba_1^2 + \mu(2a_2 - a_1^2)\right], \qquad (3)$$

where a_1 and a_2 are the invariants of strain tensor: $a_1 = u_{\alpha\beta}g^{\alpha\beta}$, $a_2 = u_{\alpha\beta}u_{\gamma\delta}g^{\alpha\gamma}g^{\beta\delta}$ ($g^{\alpha\beta}$ is the contravariant metric tensor on the unperturbed surface) and dA is the square element of a surface. The first term represents the energy of surface compression deformation while the second is due to shear deformations. The elastic force acting on the surface element is the variational derivative of this energy in respect to vector u^i.

We decompose both displacement and force vectors into normal and tangential parts. The latter can also be expressed in terms of two scalar functions, so that the displacement vector is:

$$u^i = u_n n_0^i + G^{\alpha\beta}(\partial_\alpha n_0^i)\partial_\beta u_t \;+\; G^{\alpha\beta}(\partial_\alpha n_0^i)\partial_\beta^*\tilde{u}_t, \quad (4)$$

where n_0^j is the unit vector directed along r_0^j and u_n is the amplitude of the normal displacement. Derivative $\partial_\beta^* = (1/\sqrt{G})G_{\beta\alpha}e^{\alpha\gamma}\partial_\gamma$ where $e^{\alpha\gamma}$ is a unit antisymmetric tensor and G is a determinant of the (covariant) metric tensor $G_{\alpha\beta}$ on the unit sphere. Contributions from u_t and $\tilde{u}_t$ describe the change in the surface density of the membrane and the shear deformations in tangential plane respectively. The latter part is not excited by an external flow in the regime of small deformations, therefore we will not consider its contribution in our calculations. In (4), we used decomposition near sphere but not near the capsule from at rest due to it is more convenient when the capsule form always remains nearly spherical. The components of the force vector are expressed in terms of these functions:

$$f_n = -2B(2u_n + \Delta^\perp u_t)/R, \qquad (5)$$

$$f_\alpha = \partial_\alpha[B(2u_n + \Delta^\perp u_t) + \mu(\Delta^\perp + 2)u_t]/R, \qquad (6)$$

where $\Delta^\perp$ is a Beltrami–Laplace operator on unit sphere.

In the present paper we study the dynamics of nearly spherical capsule. It is convenient to represent the position-vector of an membrane element in unperturbed state as $r_0^j = n_0^j(1 + u_{n,0})$, where $u_{n,0}$ determines the form of the capsule at rest. It is also convenient to expand all values in the series of spherical harmonics. Here we do not consider the effect of wrinkles [6], so only the contribution from the second harmonic is important to us. So we have:

$$u = u^{ij}n^i n^j. \qquad (7)$$

Matrices u_n^{ij} and u_t^{ij} are symmetric and traceless. It is assumed in (7) that n^i is a unit vector directed along the unperturbed position-vector of an membrane element $\mathcal{O}^{ij}r_0^j$ in the laboratory reference system. Thus u in (7) depends on spherical angles in laboratory reference system, in contrast with (1), (2) where ξ^α can be treated as the spherical angles in the reference system which rotates with the capsule.

We assume that the capsule is placed in an external flow $\mathbf{V}(\mathbf{r})$ with the linear profile, so that $\partial_j V_i = s_{ij} + \omega_{ij}$, where s_{ij} and ω_{ij} are constant symmetric and antisymmetric tensors respectively. We will also assume that the flow is plane which means that in a corresponding reference frame the nonzero elements of the velocity gradient matrix $\partial_j V_i$ are $\partial_y V_x = s + \omega$, $\partial_x V_y = s - \omega$. This flow can be realized in a so-called "four roll mill flow" device [20]. We consider the liquids outside and inside the capsule to have viscosities η and $\lambda\eta$ respectively, where λ is a dimensionless viscosity contrast. The mass densities of the liquids are assumed to be the same. The membrane internal viscosity is $\eta R\lambda'$, its influence was first considered in [15]. In our final calculations we consider the ratio of the elastic modules to be $B/\mu = 3$ which corresponds to the rubber shell which conserves its volume. In real experiments the ratio is $B/\mu \approx 3.7$ [1].

3. Equations which govern evolution of the capsule shell can be written in the form

$$\partial_t \hat{\mathbf{u}} = \hat{\Omega}\hat{\mathbf{u}} - \hat{\mathbf{u}}\hat{\Omega} + \mathbf{Q}\hat{s} - \frac{B}{\eta R}\check{\Gamma}\hat{\mathbf{u}}, \tag{8}$$

in terms of coefficients (7). We used the vector notation $\hat{\mathbf{u}} = \{\hat{u}_n, \hat{u}_t\}$. The derivation procedure of such equations is given in details in [13, 21, 22]. Here we present only the results, the derivation of the result will be presented later in more detailed paper.

The two first terms in the right side of equation (8) represent the contribution of the capsule rotation as a whole. The third term in the right side of (8) is from the strain components associated with s_{ij}. The forth term is due to the elastic forces and represents the relaxation of the capsule membrane to the equilibrium configuration. Such decomposition is possible because both the fluid flow and the elastic force are governed by linear equations.

Let us now make the qualitative analysis of the equation (8). The relaxation operator can be derived using the expressions for the elastic forces. Acting on the array $\hat{\mathbf{u}} = \{\hat{u}_n, \hat{u}_t\}$ this operator is the 2×2 matrix:

$$\check{\Gamma} = \frac{1}{C_1}\begin{pmatrix} 4(\lambda+16\lambda'+4) & -12(\lambda+16\lambda'+4)-24(3\lambda+4\lambda'+2)\mu/B \\ -2(7\lambda+8) & 6(7\lambda+8)+4(13\lambda+8\lambda'+12)\mu/B \end{pmatrix}, \tag{9}$$

where $C_1 = 38\lambda^2 + 150\lambda\lambda' + 89\lambda + 64\lambda'^2 + 160\lambda' + 48$. Each component of this expression is diagonal matrix 3×3 in the space of spherical harmonics with $l = 2$ [13, 15]. Operator $\mathbf{Q}$ has similar structure and is given by:

$$\mathbf{Q} = C_2/C_1\{2,1\}, \quad C_2 = 5(16 + 19\lambda + 40\lambda')/2. \tag{10}$$

It represents the action of the external stretching flow. Dimensionless surface viscosity λ' produces effect in (9), (10) similar to λ, so we put $\lambda' = 0$ below for brevity.

From the above expression one can see that the order of magnitude of the displacement vector u is determined by the parameter $\mathrm{Ca} = \eta R s/B$, when the external flow is pure elongational having $\omega = 0$. The relative strength of the rotational part of the flow is determined by another dimensionless parameter:

$$\Lambda = \frac{(1+\lambda)\eta R\omega}{B}. \tag{11}$$

Formally, definition (11) differs from the definition $\Lambda \sim \sim (1+\lambda)(\omega/s)\sqrt{\Delta}$ used in [22, 6], where Δ is the excess area parameter. In our case Δ is not constant and is a function of external flow, $\sqrt{\Delta} \sim \mathrm{Ca}$. Using the estimation for Δ in the definition for Λ [22] we arrive to (11) with the estimation. If one assumes that $\Lambda \lesssim 1$, then the magnitude of the displacement vector $u \sim \mathrm{Ca}$.

The unperturbed part of the capsule form $u_{n,0}$ undergoes rotation with the same angular velocity $\hat{\Omega}$ as deformation $\hat{\mathbf{u}}$ does in (8) due to definition (1):

$$\partial_t \hat{u}_{n,0} = \hat{\Omega}\hat{u}_{n,0} - \hat{u}_{n,0}\hat{\Omega}. \tag{12}$$

Angular velocity $\hat{\Omega}$ in (8), (12) deviates from rotation $\hat{\omega}$ imposed by the the external flow due to the capsule non-sphericity. Keeping the main contribution into $\hat{\Omega}$ in (8), one obtains that the deviation $\hat{\mathbf{u}}$ from equilibrium state trends to the stationary asymptotics

$$\hat{\mathbf{u}}_{\text{stat}} = \hat{\Gamma}_\omega^{-1}[\mathbf{Q}\hat{s}] \tag{13}$$

at large times, where action of linear operator $\hat{\Gamma}_\omega$ is determined by $\hat{\Gamma}_\omega\hat{\mathbf{u}} = \hat{\mathbf{u}}\hat{\omega} - \hat{\omega}\hat{\mathbf{u}} + \check{\Gamma}\hat{\mathbf{u}}$. These times are of the most interest from experimental point of view. The correction to angular velocity $\hat{\Omega}$ for stationary in time deformation $\hat{\mathbf{u}}$ is the same as for rigid particle [23]:

$$\hat{\Omega} = \hat{\omega} + (\hat{u}_n + \hat{u}_{n,0})\hat{s} - \hat{s}(\hat{u}_n + \hat{u}_{n,0}). \tag{14}$$

If $\partial_t\hat{\mathbf{u}}$ is not zero, r.h.s. of (14) acquires additional terms which are bilinear over $\partial_t\vec{u}$ and $\hat{u}_n$, $\hat{u}_{n,0}$.

4. Below we investigate separately the case of plane external flow in more details. The plane flow induces only harmonics which correspond to matrix elements u_n^{ij} with $\{i,j\} = \{x,y\}$. Thus $u_n(\theta,\varphi) = D\sin^2\theta\cos[2(\varphi - -\Phi)]$ in the laboratory reference system, where D is Taylor deformation parameter and Φ is the capsule inclination angle in OXY-plane.

First we investigate the capsule form when it is exactly sphere at rest. The orientation angle $\Phi = \pi/4$ if rotational part of the flow is absent, $\omega = 0$. Taylor deformation parameter is $D = (5/4)\mathrm{Ca}(3\mu^{-1} + B^{-1})$ in the case. The parameter D approaches constant limit when B increases while μ is fixed. Note however that linear theory considered here remains valid until $\mathrm{Ca} \ll (\mu/B)^2$. In the opposite limit one should take into account nonlinear (in displacement) contributions into capsule membrane force. Here we also note that the limit of locally incompressible membrane corresponds to the inequalities $\mathrm{Ca} \ll u_0$ and $\mathrm{Ca}B/\mu \sim 1$. Our theory is not valid in this limit, which was recently considered analytically in [6].

Contribution of the vorticity part $\hat{\omega}$ of the flow becomes considerable when dimensionless parameter Λ reaches values of the order of unity. The condition is equivalent to $\mathrm{Ca} \sim 1$ for shear flow and identical liquids inside and outside the capsule. One should set dimensionless parameter $(1+\lambda)\omega/s \gg 1$ in order to reach experimentally observed values of $\Lambda \sim 1$ keeping capsule form close to spherical. In what follows below we assume the viscosity contrast $\lambda \ll 1$, since the limit corresponds to experimental situation [1, 3]. Taylor deformation parameter is

$$D_{\mathrm{stat}} = \frac{25\mathrm{Ca}}{2}\sqrt{\frac{(36/25)\Lambda^2+1}{324\Lambda^4+189\Lambda^2+1}} \qquad (15)$$

in this limit. At large $\Lambda \gg 1$ Taylor deformation parameter approaches the value $D_{\mathrm{stat}} \to C_2/C_1$, which does not depend on the capsule membrane properties but only on the external flow geometry and the viscosity contrast λ. The limit corresponds to liquid drop with absence of surface tension, the motion of a drop immersed in solvent was considered in [24]. Law (15) is graphically depicted on Fig. 1 in appropriate variables. The capsule inclination angle

$$\Phi_{\mathrm{stat}} = \frac{1}{2}\arctan\left[\frac{5}{\Lambda\,(108\Lambda^2+69)}\right] \qquad (16)$$

for $\lambda \to 0$. The orientation angle approaches zero, $\Phi_{\mathrm{stat}} \to 0.023/\Lambda^3$ when $\Lambda \gg 1$.

Let us now analyze the case when the capsule is not purely spherical at rest. The deviation of the capsule form from the sphere is described now by sum of functions $u_{n,\mathrm{stat}} + u_{n,0}$, the first one being stationary in time, see Eq. (13), while the second one oscillating with angular velocity ω. We parametrize equilibrium shape of capsule as $u_{n,0} = D_0 \sin^2\theta\cos[2(\varphi - \Phi_0)]$, where $\Phi_0 = \omega t$. The influence of contribution to $u_{n,0}$ from three others harmonics with full angular momentum be equal to $l = 2$ will be discussed later. Thus the capsule never achieves stationary form. The capsule is visually in tumbling regime if $D_{\mathrm{stat}} < D_0$, and Taylor parameter D changes during the period of tumbling. The capsule is in trembling (or in tank-treading with oscillations) if $D_{\mathrm{stat}} > D_0$, Taylor deformation parameter is $D = \{D_{\mathrm{stat}}^2 + D_0^2 + 2D_{\mathrm{stat}}D_0\cos[2(\Phi_{\mathrm{stat}} - \Phi_0)]\}^{1/2}$. In particular, amplitude of oscillations of D is constant and equal to D_0, that corresponds to experimental observations [1]. The inclination angle Φ in OXY-plane is determined by equation $\sin[2(\Phi - \Phi_{\mathrm{stat}})) = (D_0/D)\sin[2(\Phi_{\mathrm{stat}} - \Phi_0)]$.

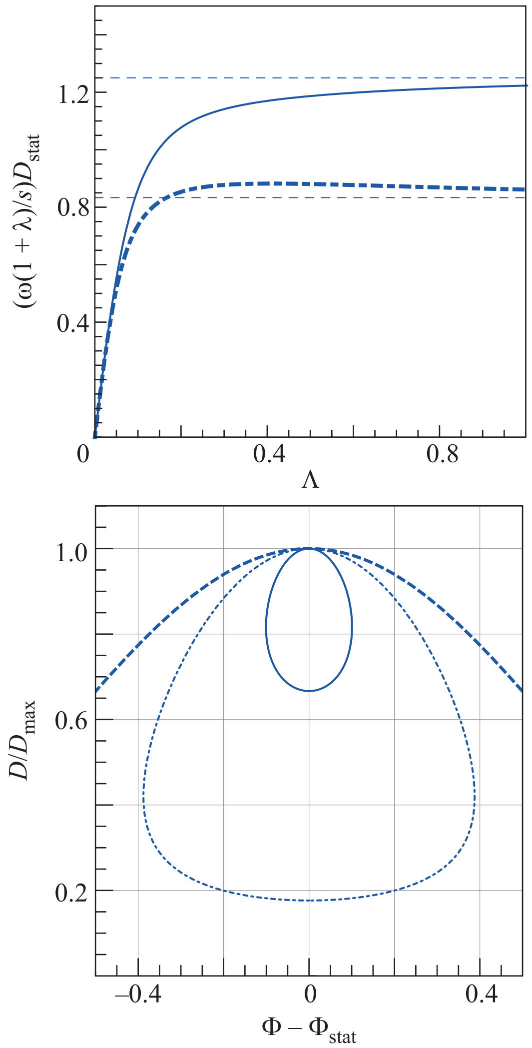

Fig. 1. On the top: dependence of Taylor deformation parameter D_{stat} on s and ω at small λ (dotted line) and large $\lambda \gg 1$ (solid line), plotted in appropriate axes for $(1+\lambda)\omega/s \gg 1$. On the bottom: correlated oscillations of capsule form and inclination angle, where Φ is measured in radians, for D_0/D_{stat} be equal to 0.2 (solid line), 0.7 (dashed-solid line) and 1.3 (dashed line)

Joint dynamics of D and Φ is depicted on Fig. 1. Tumbling occurs at weak external flows, the corresponding lines are not closed on D–Φ-plane. At stronger external flows the capsule is in trembling regime, and the lines become closed.

Two dimensionless parameters D_0/Ca and Λ fully determine the type of the capsule dynamical regime at the limit of small or large viscosity contrast λ. Thus it is possible to plot a two-dimensional phase diagram for capsule dynamical regimes, as it was done for vesicles [18]. It is depicted on Fig. 2, where Λ is substituted by its algebraic combination in order to transition lines

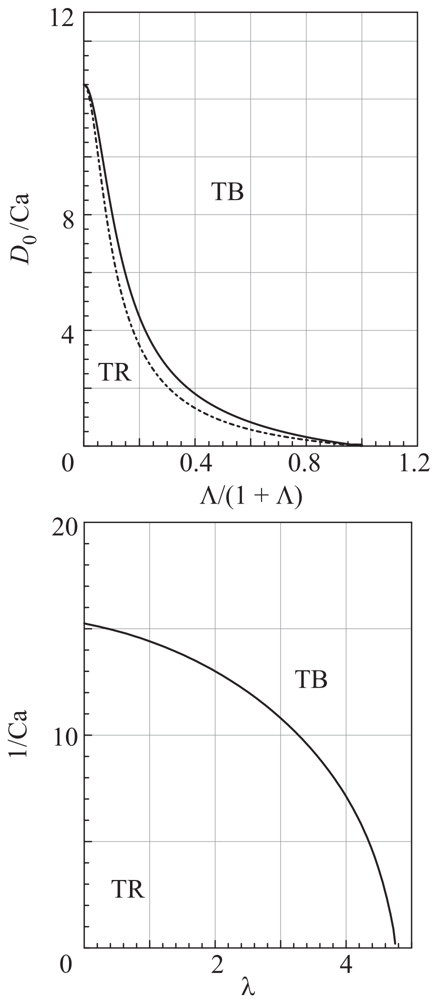

Fig.2. On the top: phase diagram for capsule dynamic regimes plotted in universal dimensionless variables. Solid line is for limit $\lambda \gg 1$, dashed – for $\lambda \ll 1$. Lines correspond to transition from tumbling, which is under line, to trembling, which is realized above line. On the bottom: phase diagram plotted for shear flow $s = \omega = \dot{\gamma}$. Taylor deformation parameter at the rest is $D_0 = 0.2$

between trebling and tumbling have finite length. Note, that the form of the curve changes if ratio B/μ changes.

We also plotted phase diagram in the form used in [16] for particular case of shear flow, it is depicted on Fig. 2 on the bottom. On the plane the position of the transition line between trembling and tumbling is not invariant to Taylor deformation parameter at rest D_0.

It is reported in experimental work [1] about linear dependence of angular velocity Ω of the capsule and Taylor deformation parameter D at $\mathrm{Ca} \gg D_0$ in shear flow. It follows from (13), (14) that the correction to angular velocity is

$$\Omega/\omega - 1 = -(5/4)\mathrm{Ca}^2(276 + 209\lambda + 220\lambda'). \qquad (17)$$

Nonlinear in flow strength correction to the deformation amplitude is $D/(25\mathrm{Ca}/2) - 1 = -94\Lambda^2$ from (15), and the correction does not coincide with correction to angular velocity (17) at limit $\lambda, \lambda' \to 0$. In general case, when $(1+\lambda)\omega/s \lesssim 1$, one needs to account non-quadratic corrections in deformation for elastic energy to find the correction to D. These corrections also determine ultimate orientation of the capsule as a whole, since they govern rotation of matrix $\hat{u}_{0,n}$ around an axis lying in plane OXY. The rotation leads to alternation in D_0 since it is determined as a Taylor parameter for the capsule cross section in OXY plane.

5. We have considered dynamics of a micro-capsule with compressible membrane placed into external flow particularly interesting in the case of the planar flow. The form of the capsule is assumed to be near-spherical and the membrane forces were calculated in the linear approximation in the membrane displacement. We have established that the capsule dynamics can be described by two dimensionless parameters in the limits of high and low viscosity contrast between the fluid inside the capsule and the solvent. One of the parameters describes the relative strength of the rotational part of the external flow and the other the ratio of the capillary number and the Taylor deformation parameter of the capsule at rest.

We analytically found the capsule deformation as a function of these two dimensionless parameters, see Fig. 1, and plotted the phase diagram of the capsule dynamical regimes, see Fig. 2. Universal dependence of such kind is interesting from experimental point of view since it allows one to cover full plane of the diagram by changing the appropriate physical parameters, as it was done recently in [19].

It is interesting to compare our results with those reported in [6, 8] concerning capsules with incompressible membrane. The general properties of the phase diagram obtained from [6, 8] and in agreement with those found by us. Particularly, capsule is in trembling (or vacillating-breathing) regime at high Ca number, and in tumbling regime if the number is small. Transition from trembling to tumbling occurs when the viscosity contrast increases. The transition driving by the viscosity contrast and the strength of the rotational part of the external flow is similar to vesicle dynamics [22]. On the contrary, the transition caused by capillary number Ca is specific for capsules. The position of the transition line is determined by the requirement that the deformation caused by the external flow has the same magnitude as the initial non-sphericity of the capsule.

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