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# Hydrodynamic forces acting on a microscopic emulsion drop growing at a capillary tip in relation to the process of membrane emulsification

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**Abstract**

Here, we calculate the hydrodynamic ejection force acting on a microscopic emulsion drop, which is continuously growing at a capillary tip. This force could cause drop detachment in the processes of membrane and microchannel emulsification, and affect the size of the released drops. The micrometer-sized drops are not deformed by gravity and their formation happens at small Reynolds numbers despite the fact that the typical period of drop generation is of the order of 0.1 s. Under such conditions, the flow of the disperse phase through the capillary, as it inflates the droplet, engenders a hydrodynamic force, which has a predominantly viscous (rather than inertial) origin. The hydrodynamic boundary problem is solved numerically, by using appropriate curvilinear coordinates. The spatial distributions of the stream function and the velocity components are computed. The hydrodynamic force acting on the drop is expressed in terms of three universal functions of the ratio of the pore and drop radii. These functions are computed numerically. Interpolation formulas are obtained for their easier calculation. It turns out that the increase in the viscosity of each of the two liquid phases increases the total ejection force. The results could find applications for the interpretation and prediction of the effect of hydrodynamic factors on the drop size in membrane emulsification.


*Keywords:* Membrane emulsification; Microchannel emulsification; Hydrodynamic forces; Drop formation at capillary tip

## 1. Introduction

The method of membrane emulsification attracted a considerable interest and found numerous applications during the last decade. The method was applied in many fields, in which monodisperse emulsions are needed. In food industry it was used for production of oil-in-water (O/W) emulsions: dressings, artificial milk, cream liqueurs, as well as for preparation of some water-in-oil (W/O) emulsions: margarine and low-fat spreads. Another application of this method is for fabrication of monodisperse colloidal particles: silica–hydrogel and polymer microspheres; porous and cross-linked polymer particles; microspheres containing carbon black for toners, etc. A third field is the production of multiple emulsions and microcapsules, which have found applications in pharmacy and chemotherapy. Detailed reviews could be found in Refs. [1–5]. Closely related to the membrane emulsification is the method employing capillary tubes or microchannels to produce monodisperse emulsions [6–9].

The key problem of membrane emulsification is related to the explanation and prediction of the dependence of the drop size on the system's parameters: pore diameter; flux of the disperse phase along the pores; applied cross flow in the continuous phase; viscosity of the oil and water phases; interfacial tension and kinetics of surfactant adsorption, etc. (Here and hereafter we call "disperse" the phase from which the drops are made, despite the fact that this phase is continuous before the drop detachment from the membrane.) Different approaches have been used to solve this problem: by regression analysis of experimental data [9]; by modeling of the drop expansion and surfactant adsorption by surface evolver [10,11]; by three-dimensional computational fluid dynamics simulations [12,13], and by lattice Boltzmann simulations [14]. The quantitative the-

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oretical analysis demands one to determine the forces exerted on the growing emulsion drop and to establish the mechanism of drop detachment from the pores.

In some experiments, cross flow is applied in the continuous phase parallel to the membrane surface. It gives rise to *drag* and *lift* hydrodynamic forces that tilt the protruding drop and help for its detachment from the membrane [4,14,15]. In addition, the flow of the disperse phase through the capillary, as it inflates the droplet, engenders a hydrodynamic *ejection* force that also tends to detach the droplet from the pore [15].

It is possible to produce monodisperse emulsions by membrane emulsification in the absence of cross flow in the outer liquid phase [3]. In this case, the drag and lift hydrodynamic forces are missing, but the ejection force alone is able to detach the drops from the pores. The hydrodynamic estimates (see Section 3 below) show that under typical experimental conditions this process happens at small Reynolds numbers, for which the inertial terms in the Navier–Stokes equation are negligible. Hence, under such conditions the ejection force has a predominantly viscous (rather than inertial) character.

The full hydrodynamic problem for detachment of emulsion drops in cross flow is rather complicated. It could be split into two separate problems: (i) calculation of the ejection force in the absence of cross flow in the outer liquid, and (ii) accounting for the effect of the cross flow. Our goal in the present article is to solve the first problem.

The paper is organized as follows. In Section 2, we consider the kinematics of drop expansion. In Section 3, the basic equations and boundary conditions are formulated. In Section 4, appropriate curvilinear coordinates are introduced to transform the three physical domains (capillary channel, drop and outer phase) into rectangles. The hydrodynamic boundary problem is solved numerically and the velocity field is calculated. Finally, in Section 5 the hydrodynamic force acting on the emulsion drop is computed and interpolation formulas are obtained for its easier calculation. Details of the theoretical derivations are given as appendices to this paper—see Supplementary material.

## 2. Kinematics of drop expansion

We consider the expansion of an emulsion drop, which is growing at the tip of a capillary. Our purpose is to quantify the hydrodynamic forces acting on the drops, which are formed at the openings of the pores of an emulsification membrane. We are dealing with microscopic drops, for which the gravitational deformation of the drop is negligible. Here, we consider the simpler case, in which there is no cross-flow in the outer liquid phase; i.e., the only motion in the outer liquid is due to its displacement by the growing drop.

Because we are dealing with small drops, we will simplify our treatment by the assumption that the drop surface is (approximately) spherical. The membrane pore will be modeled as a cylindrical channel, see Fig. 1. We will denote the radius of the drop surface by $R_s$, and the inner radius of the pore—by $R_p$. To describe the process of drop formation, we will use cylindrical coordinates $(r, z)$, where the $z$-axis coincides with the axis

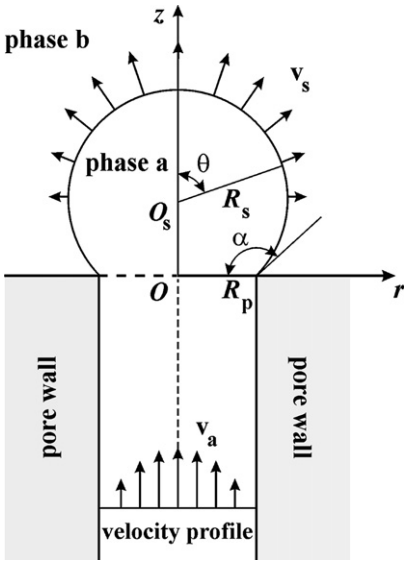


Fig. 1. Drop from the liquid phase 'a' growing at the orifice of a membrane pore. Phase 'b' is the outer liquid medium. $R_p$ and $R_s$ are, respectively, the radii of the cylindrical pore and spherical drop surface. The "protrusion" angle $\alpha$ characterizes the size of the drop ($0 \leqslant \alpha \leqslant 180^\circ$), whereas the angle $\theta$ characterizes the positions of the material points at the drop surface; $\mathbf{v}_s$ is the surface velocity.

of rotational symmetry of the system, and the plane $z = 0$ coincides with the outer membrane surface (Fig. 1).

The inner and outer liquids will be referred as "phase a" and "phase b," respectively. For example, "phase a" could be oil and "phase b"—water, or vice versa. Due to the symmetry, the velocity fields, $\mathbf{v}_a$ and $\mathbf{v}_b$, in the respective phases can be expressed in the form:

$$\mathbf{v}_a = u_a \mathbf{e}_r + w_a \mathbf{e}_z, \qquad \mathbf{v}_b = u_b \mathbf{e}_r + w_b \mathbf{e}_z, \tag{2.1}$$

where $\mathbf{e}_r$ and $\mathbf{e}_z$ are the unit vectors of the $r$- and $z$-axes. Inside the cylindrical channel, far from its orifice, we have Poiseuille flow of the inner liquid [16]:

$$u_a = 0, \qquad w_a = 2v_m\left(1 - \frac{r^2}{R_p^2}\right) \quad \text{for } 0 \leqslant r \leqslant R_p \text{ and } z \to -\infty. \tag{2.2}$$

Here $v_m$ is the mean velocity, and the subscript "a" denotes the inner liquid phase. The flow rate, $Q$, of the inner liquid is

$$Q = \pi R_p^2 v_m = \frac{dV}{dt}, \tag{2.3}$$

where $V$ is the volume of the growing drop, and $t$ is time. The volume can be expressed in the form:

$$V = \frac{\pi R_p^3}{3} \frac{2 + \cos\alpha}{(1 + \cos\alpha)^2} \sin\alpha, \tag{2.4}$$

where the angle $\alpha$ characterizes the protrusion of the droplet from the pore (Fig. 1), and will be termed below "protrusion angle." The differentiation of Eq. (2.4), in view of Eq. (2.3), yields:

$$\frac{d\alpha}{dt} = \frac{v_m}{R_p}(1 + \cos\alpha)^2. \tag{2.5}$$