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Asymmetric rheological behaviors of double-emulsion globules with asymmetric internal structures in modest extensional flows

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A B S T R A C T

The oriented shift and inverse of double-emulsion globules containing two inner droplets with different sizes and locations in modest extensional flows are investigated numerically in this paper through a boundary element method. The asymmetric layout of daughter droplets leads to the asymmetric inner flow field and pressure field inside the globule, which causes its asymmetric rheological behaviors. The direction of shift is determined not only by the size ratio r_R^2/r_L^2 and the location ratio d_R/d_L of two inner droplets, but also by some flow factors such as the capillary number Ca. There is a critical capillary number Ca_c as a function of r_R^2/r_L^2 and d_R/d_L, beyond which the globule will move to the right, otherwise, it will shift to the opposite direction.

1. Introduction

Multiple emulsions are complex soft globules which have skillfully designed internal structures probably including multiple layers and multiple engulfed droplets in each layer [1]. They have drawn much attention recently in different disciplines as they have broad potentials in drug delivery, cosmetics, food industry, and so on [2]. Generally, these complex globules are delivered through flows, and naturally their rheological behaviors are important and necessary to be investigated carefully.

The rheological behaviors of multiple-emulsion globules in various flow patterns have been studied experimentally and numerically [3–18]. It was investigated and disclosed experimentally that the double-emulsion globules under the flow shear could break up controllably and release their inclusions when they were squeezed through contractions with different sizes [3,4]. Besides experiments, the deformation and breakup of multiple-emulsion globules have also been investigated through numerical simulations. Especially for the rheological behaviors of concentric double emulsions (CDE) in symmetric extensional and shear flows, they have been studied widely and carefully [5–10]. The deformation and breakup of CDE in infinite extensional flows was investigated by Stone and Leal in 1990 [5]. Two different breakup mechanisms of the globule, (1) the continuous elongation without steady shapes, and (2) the contact of inner and outer interfaces, were presented by them. Smith et al. [8] produced a phase diagram to show the morphologies of no breakup and breakup with daughter droplets of different numbers when they investigated the stretch and relaxation of CDE in shear flows. In addition, several research groups have also studied numerically the translation of the multiple-emulsion globule in circular or constriction tubes [11–14]. Wang et al. [14] disclosed that a globule with two unequal inner droplets is easier to pass the constriction when its smaller inner droplet locates in its rear initially during their investigation of the transition of this type of globule in an axisymmetric microfludic constriction.

Recently, several efforts have been done to study the effects of the asymmetric internal structure of multiple emulsions on the rheological behaviors of the globules and their inner droplets. Qu et al. [15] investigated an eccentric compound droplet in extensional flows and presented that the droplet tended to shift away from its original location because of the asymmetric layout of the inner droplet. The globule always shifts in the same direction with the daughter droplet and its speed is proportional to the distance between the inner droplet and the center of the outer droplet. Wang and coworkers [16–18] have designed a type of complex asymmetric globules of multiple emulsions which contain three layers (one inner droplet in the 2nd layer and three or two in the 3rd layer). Due to the asymmetric layout of the grand-daughter droplets (GDD), the daughter droplet (DD) will move in an oriented direction when the globule is in a symmetric extensional flow. Especially, Wang et al. [18] presented that both the structural asymmetry parameter A_s and the capillary number Ca affect the shift direction, and changing these factors might result in the reverse of the shift. However, in order to define a monotonically increasing A_s, the locations of GDDs are fixed. They did not investigate the effect of location changes of GDDs.

Meanwhile, Wang and coworkers [19,20] investigated the rheological behaviors of a simple droplet in axisymmetric extensional flows (asymmetric flow fields from left to right), in which the asymmetries of the flow systems are generated from the physical factors outside the

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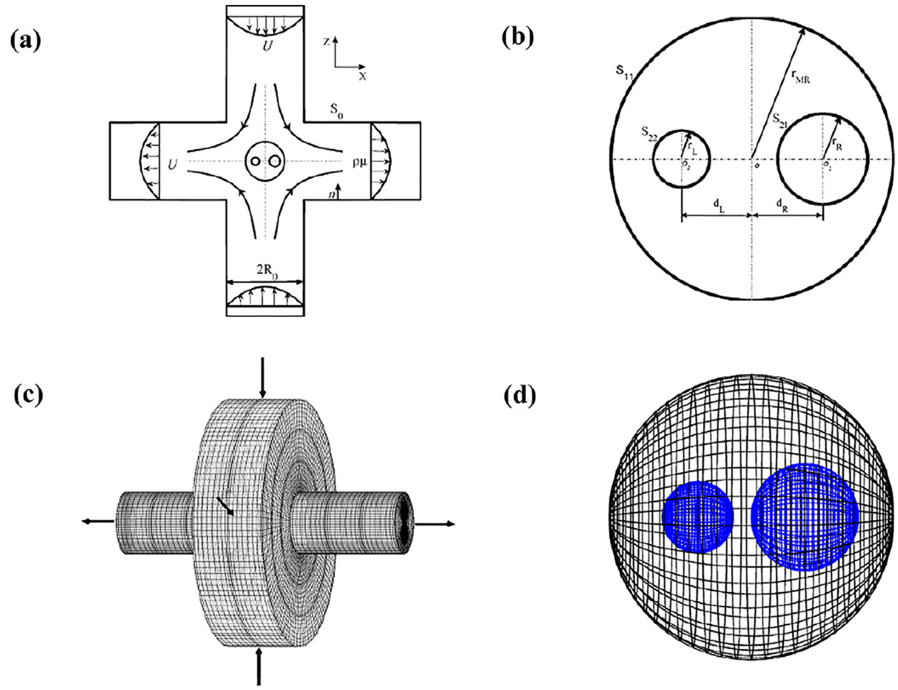

Fig. 1. The illustration of the 2-dimensional cross-like microfluidic device (a) and the 2-dimensional multiple-emulsion globule (b). The illustration of the equivalent axisymmetric cross-like microfluidic device (c) and the equivalent asymmetric multiple-emulsion globule (d).

droplet. They reported that the droplet will not break up and leave from one side of the axisymmetric device for a small Ca. When the asymmetry is fixed, the move direction is always the same for a simple droplet. For a multiple-emulsion globule with asymmetric inner structures in an axisymmetric extensional flow (also symmetric from left to right), as the asymmetry of the entire flow system is generated from the physical factor inside the mother droplet, the phenomenon under modest capillary numbers might be different.

2. Mathematical formulation and numerical method

In this paper, the oriented shift of a double-emulsion globule with two unequal daughter droplets (DD) in an axisymmetric cross-like microfluidic device (Fig. 1) is investigated numerically. In Fig. 1(b), the interface of the globule is S_{11}, and the interfaces of the two inner droplets are S_{21} and S_{22}, respectively. In order to find out factors affecting the oriented shift, three important parameters will be investigated. The first one is the capillary number Ca $= \mu U/\gamma$, where $U = 1$ is the average velocity of the continuous phase (CP) at inlets, $\mu = 1$ is the viscosity of CP, and γ is the interfacial tension whose variation will change Ca. The other two are dimensionless numbers to characterize the asymmetry (see Fig. 1(b)), which are the size ratio of two DDs ${r_R}^2/{r_L}^2$ and the location ratio d_R/d_L. Here, r_R and r_L are the radii of the right and left DD, d_R and d_L are the eccentric distances of the right and left DD, respectively. The volume ratio Φ of the total size of two DDs to that of the mother droplet is 13.6%, and the viscosity ratio λ (the dispersed phase to CP) is fixed to 1.2. Initially, the globule and two DDs are both spherical and stationary. The radius of the globule r_{MR} is $0.5R_0$, and the radii of two DDs will change in our study. Initially, two DDs are both located on x-axis, and d_R and d_L will be changed.

The problem studied here is about the rheological behaviors of asymmetric double-emulsion globules in micro-channels (Fig. 1). The width of the microchannel and the radius of the droplet are generally in the micron scale (10^{-6} m). Thus, when the density of CP and the shear rate are not very large, generally, Reynolds number Re is always less than unit. Thus, this hydrodynamic problem is presumed in the Stokes regime.

Eqs. (1) and (2) are the continuity equation and the Stokes equations, respectively. They are the governing equations of the system shown in Fig. 1(a) and (b).

$$\nabla \cdot \boldsymbol{u} = 0, \tag{1}$$

$$-\nabla P + \mu \nabla^2 \boldsymbol{u} = 0. \tag{2}$$

Here $\boldsymbol{u}$ is the velocity of the continuous phase, P is the dynamic pressure, and μ is the viscosity of the continuous phase. These two equations could be employed both for the continuous phase and for all droplets of the double emulsions. Certainly, for different droplets, physical parameters of the equations ought to be replaced by those of droplets.

The velocity at a point $\boldsymbol{x}_0$ on all boundaries including S_0 (the wall, inlet and outlet), and the droplet surfaces (S_{11}, S_{21} and S_{22}) could be calculated by the boundary integral equation (BIE),

$$\begin{aligned} LHS = &-\int_{S_0}\left[S\cdot \boldsymbol{f} - \mu \boldsymbol{T}\cdot \boldsymbol{u}\cdot \boldsymbol{n}\right] dS \\ &-\int_{S_{1,1}}\left[S\cdot \Delta f_{1,1} - (1-\lambda_{1,1})\mu T\cdot u\cdot n\right] dS \\ &-\int_{S_{2,1}}\left[S\cdot \Delta f_{2,1} - (\lambda_{1,1}-\lambda_{2,1})\mu T\cdot u\cdot n\right] dS \\ &-\int_{S_{2,2}}\left[S\cdot \Delta f_{2,2} - (\lambda_{1,1}-\lambda_{2,2})\mu T\cdot u\cdot n\right] dS \end{aligned} \tag{3}$$

where LHS is

$$LHS = \begin{cases} 2\pi\mu \boldsymbol{u}(\boldsymbol{x}_0) & \boldsymbol{x}_0 \in S_0 \\ 2\pi\mu(1+\lambda_{1,1})\boldsymbol{u}(\boldsymbol{x}_0) & \boldsymbol{x}_0 \in S_{1,1} \\ 2\pi\mu(\lambda_{1,1}+\lambda_{2,1})\boldsymbol{u}(\boldsymbol{x}_0) & \boldsymbol{x}_0 \in S_{2,1} \\ 2\pi\mu(\lambda_{1,1}+\lambda_{2,2})\boldsymbol{u}(\boldsymbol{x}_0) & \boldsymbol{x}_0 \in S_{2,2} \end{cases}, \tag{4}$$

where $\boldsymbol{S}$ is the fundamental solution for Stokes equations, and $\boldsymbol{T}$ is the stress kernel.