Formation of microdroplets in Newtonian and shear thinning fluids flowing in coaxial capillaries: Numerical modeling

S A Vagner\textsuperscript{1}, S A Patlazhan\textsuperscript{1,2} and C A Serra\textsuperscript{3}

\textsuperscript{1} Institute of Problems of Chemical Physics of the Russian Academy of Sciences, Academician Semenov Avenue 1, Chernogolovka, Moscow Region 142432, Russia
\textsuperscript{2} Semenov Institute of Chemical Physics of the Russian Academy of Sciences, Kosygina 4, Moscow 119991, Russia
\textsuperscript{3} Universite de Strasbourg, 4 rue Blaise Pascal, Strasbourg 67081, France

E-mail: sapat@yandex.ru

Abstract. The peculiarities of the Newtonian droplets formation in a Newtonian or non-Newtonian fluid from coaxial capillaries are studied by means of numerical modeling. As a non-Newtonian medium, a shear thinning fluid is considered whose viscosity decreases with the increase of the shear rate according to the Carreau-Yasuda rheological model. It is shown that in the case of a Newtonian continuous fluid, the calculated sizes of the droplets decrease with an increase of the ratio of the flow rates in the outer and inner capillaries which is in a good agreement with experimental data. On the other hand, for the shear thinning continuous medium, droplet sizes become significantly larger than those for the Newtonian fluid. Besides, in the latter case, the droplet sizes weakly depend on the ratio of flow rates of the continuous and dispersed fluids.

1. Introduction

Microscopic drops are of great interest for various physical, medical and biological applications. They are used as chemical microreactors [1, 2], microcontainers for targeted drug delivery [3, 4], microcells for photonic and phonon crystals [5], etc. One of the key problems is the capability of generating monodisperse microdroplets of a given size. To solve this problem, various microfluidic devices have been designed [6–8] since sizes of droplets are mostly determined by cross-sections of microchannels and flow rates of continuous and dispersed phases. Among these devices, the coaxial capillary flow belongs to the simplest and productive method of generation of monodisperse microdroplets [9,10]. The droplet formation occurs due to their detachment or disintegration of jet of the dispersed fluid from the internal capillary due to the interaction with the incompatible continuous medium flowing through the external capillary (figure 1).

At present, the droplet formation in the coaxial capillaries was studied solely for the Newtonian fluids. The question of the influence of non-Newtonian continuous medium remained open. However, this problem deserves special attention because polymer solutions are used often in microfluidic technologies. Considering that viscosity of such non-Newtonian fluids depends on the shear rate, it is clear that it should influence the sizes of the generated microdroplets. This problem is of fundamental interest since it requires an understanding of the relationship...
between the dynamics of droplet formation with the evolution of the viscosity distribution of non-Newtonian fluid around the droplet.

2. Model
The 3D flow of two incompatible fluids in the coaxial capillaries shown in figure 2 is considered. The inner diameter of the external capillary is $d_1$ while sizes of the inner capillary are described by the internal $d_{2,\text{int}}$ and outer $d_{2,\text{out}}$ diameters. The flow rates and viscosities of the fluids in the outer and inner capillaries are $Q_c$, $\eta_c$ and $Q_d$, $\eta_d$, respectively, while their densities are taken to be equal $\rho_c = \rho_d = \rho$. The interfacial tension between the fluids is $\sigma$. The droplets formation depends on the following dimensionless parameters: (i) the ratio of the flow rates $q = Q_c/Q_d$, (ii) viscosity ratio $m = \eta_c/\eta_d$, and (iii) capillary numbers $Ca_c = \eta_c V_c/\sigma$ and $Ca_d = \eta_d V_d/\sigma$ where $V_c$ and $V_d$ are the average velocities of the fluids in the outer and inner capillaries, respectively.

The flow of the incompressible fluids obeys the following Navier–Stokes equations and incompressibility condition:

$$
\rho \left( \frac{\partial u_c}{\partial t} + (u_c \cdot \nabla) u_c \right) = -\nabla p_c + \nabla \left( \eta_c (\nabla u_c + (\nabla u_c)^T) \right),
$$

$$
\rho \left( \frac{\partial u_d}{\partial t} + (u_d \cdot \nabla) u_d \right) = -\nabla p_d + \eta_d \Delta u_d,
$$

$$
\nabla \cdot u = 0.
$$

Here $u_c$, $p_c$ and $u_d$, $p_d$ are the local velocities and pressures of the continuous and dispersed fluids, respectively; index T is the matrix transpose operator. The continuity condition of the flow velocity is applied at the interfaces between the fluids while the velocity is zero on the capillary walls (stick boundary conditions). In equation (2) we took into account that the dispersed fluid $\eta_d$ is the Newtonian one with a constant viscosity while viscosity of the non-Newtonian continuous medium $\eta_c$ may be variable within flowing two-phase system and in the vicinity of the inner capillary. Viscosity of inelastic shear thinning fluid is described in terms of the Carreau–Yasuda model [11].
Figure 3. The dynamic adaptive mesh of high resolution at the interface between the fluids.

For a simple shear flow of a homogeneous medium it is described as
\[
\eta(\dot{\gamma}) = \eta_{\infty} + (\eta_0 - \eta_{\infty})[1 + (\lambda \dot{\gamma})^a]^{(n-1)/a},
\]
where \( \dot{\gamma} \) is the shear rate, \( \eta_0 \) and \( \eta_{\infty} \) are the medium viscosities at zero and infinite shear rate, respectively. The shear thinning fluids are characterized by \( n < 1 \). As an example, in this paper we consider an aqueous solution of polyacrylamide with a polymer concentration of 500 ppm which is described by the following parameters: \( \eta_0 = 1.08 \text{ Pa}s, \eta_{\infty} = 0.0023 \text{ Pa}s, \lambda = 5 \text{ s}, n = 0.2 \) and \( a = 1 \) [12]. In the case of the heterogeneous two-phase system, the local strain rate tensor \( \mathbf{D}_c = \frac{1}{2}(\nabla \mathbf{u}_c + (\nabla \mathbf{u}_c)^T) \) of the non-Newtonian continuous phase should be taken into account. In this case, the viscosity of the Carreau–Yasuda fluid is described in a more general form
\[
\eta(D) = \eta_{\infty} + (\eta_0 - \eta_{\infty})\left[1 + (2\lambda^2 I_2)^{a/2}\right]^{(n-1)/a},
\]
where \( I_2 = \mathbf{D}_c : \mathbf{D}_c \) is the second invariant of the strain rate tensor.

The Navier–Stokes equations (2) have been solved numerically on the basis of the Computational fluid dynamic package OpenFoam [13]. The finite-volume method was applied for the numerical integration of the differential equations. Separation of velocity and pressure fields was carried out with use of the PISO (pressure-implicit with splitting operator) algorithm. For numerical calculations the 3D mesh was adapted dynamically in such a way to correspond to the moving interface to keep the highest resolution (figure 3). This approach allows significant reduction of the computing time and a high accuracy in calculation of capillary forces at the fluid interface.

3. Results and discussion

Before proceeding the analysis of the effect of a non-Newtonian continuous phase, we have considered first the capability of the computational model to describe the process of droplet formation within a Newtonian fluid and compared the results of numerical calculations with experimental data obtained in work [10] with the following diameters of the capillaries: \( d_1 = 1.6 \text{ mm}, d_{2,\text{int}} = 260 \mu\text{m} \) and \( d_{2,\text{out}} = 514.4 \mu\text{m} \). Viscosities of the continuous and dispersed fluids were set as \( \eta_c = 0.35 \) and \( \eta_d = 7.2 \times 10^{-4} \text{ Pa}s \), respectively. Figure 4 presents the calculated and experimental images of shapes of the droplet during its outflow for the following flow rates of the dispersed and continuous fluids: \( Q_d = 5 \mu\text{L/min} \) and \( Q_c = 150 \mu\text{L/min} \). The interfacial tension is equal to \( \sigma = 3.3 \times 10^{-3} \text{ N/m} \). It is seen that the calculated shapes of the droplet at different instants well agree with the experimental observations [10]. The comparison of the calculated and experimental droplet sizes at different values of the flow rate ratio \( Q_c/Q_d \) is presented in figure 5 for the same dispersed liquid as in figure 4 and two Newtonian continuous fluids having
Figure 4. Dynamics of the droplet formation: calculation (a) and experiment [10] (b).

Figure 5. The calculated (solid lines) and experimental [10] (dashed lines) dependences of the droplet diameter versus the flow rate ratio at $\eta_c = 0.8$ Pa s (solid circles—calculation; open circles—experiment) and $\eta_c = 0.35$ Pa s (solid squares—calculation; open squares—experiment).

viscosities $\eta_c = 0.35$ and $\eta_c = 0.8$ Pa s as in work [10]. It is seen that the calculated results are in a good agreement with the experimental data. In particular, for a Newtonian continuous fluid, the size of the droplets formed decreases when $Q_c/Q_d$ or viscosity of the continuous phase are increased. Considering that the size of the droplets results from the competition between the shear stress imposed by the flow of the continuous phase and the interfacial tension [10], these observations result from an increase in the shear stress due to the growth of local shear rates. The maximum droplet size approximately corresponds to the outer diameter of the inner capillary $d_{2,\text{out}}$. The obtained results indicate the reliability of the numerical model.

Figure 6 shows the numerical snapshots of the formation process of the Newtonian droplets for the same dimensions of the capillaries but two different types of continuous phases: (i) a Newtonian fluid with the given viscosity of $\eta_c = 1.08$ Pa s (figure 6(a)) and (ii) a shear thinning medium (figure 6(b)) whose shear rate dependent viscosity is described by equation (3) corresponding to the 500 ppm aqueous solution of polyacrylamide (see above). Note that the zero shear rate viscosity of this non-Newtonian fluid coincides with the viscosity of the Newtonian continuous fluid. In the figures, the ratio of the capillary flow rates is taken to equal
Figure 6. Snapshot of Newtonian droplet formation in the coaxial capillaries with flowing Newtonian (a) and 500 ppm aqueous solution of polyacrylamide (b) continuous fluids at $Q_c/Q_d = 30$.

Figure 7. Droplet diameter versus flow rate ratio for shear thinning (solid triangles) and Newtonian (open triangles) continuous fluids.

to $Q_c/Q_d = 30$. It can be seen that the size of the droplet formed in the shear thinning fluid is substantially larger than that in the Newtonian continuous phase.

Figure 7 shows dependences of the Newtonian droplet diameter formed in both Newtonian and shear thinning continuous fluids on the flow rate ratio. It follows that the droplets are significantly larger while they are formed in the shear thinning solution than in Newtonian continuous liquid in the whole range of $Q_c/Q_d$ values. It follows from the numerical calculations that in contrast to Newtonian fluids, sizes of droplets formed in the flowing shear thinning medium are almost independent of the flow rates ratio.

To understand the effect of the growing size of a droplet forming in the flowing shear thinning fluid, the viscosity field near the droplet was visualized. The area of the reduced viscosity around the droplet is observed in figure 8(a). This results in a notable decrease in viscous stresses. On the other hand, an area of the increased viscosity is formed just in front of the droplet. Both of these factors prevent the early detachment of the droplet and favor to significant growth of its size. On the contrary, viscosity of the Newtonian continuous medium is kept constant being equal to the largest zero viscosity of the non-Newtonian medium (figure 8(b)). As a result, the Newtonian fluid accumulates larger stresses leading to the smaller droplets.
4. Conclusions
The numerical modeling of the formation of a Newtonian droplet from coaxial capillaries under the flow of either a Newtonian or a shear thinning continuous fluids has been carried out. It was found that the droplets formed in the flowing inelastic shear thinning fluid are much larger than those created in the Newtonian continuous phase. This effect is associated with the areas of the reduced and increased viscosity forming in the flowing shear thinning fluid upstream and downstream of the droplet. In contrast to the case of a Newtonian continuous fluid, the size of the droplets formed in the non-Newtonian medium is almost independent of the flow rate ratio in the capillaries.

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