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## Cox modified model to describe the time evolution of a drop deformation for high viscosity drops in simple shear flow

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## ABSTRACT

In this work a modification of Cox's model for the deformation of a highly viscous drop is presented, which describes accurately the period of the oscillations during transient deformations. It is demonstrated that the oscillation period depends not only on the shear rate as predicted by Cox model but also on the viscosity ratio. This modified model is obtained from the analysis of numerical simulations of drop deformation using the boundary element method, while the experimental results are those obtained in quasi-simple shear flow generated in a two roll mill (2TRMs) device, as well as experimental data available in the literature. The predictions of the periods match very well with experimental data.

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## 1. Introduction

The deformation and breakup of a droplet embedded in a continuous immiscible liquid phase subjected to linear flows has been of interest due to the large set of applications dealing with drops dispersed in a fluid matrix. Thus, an extensive data set has been published, and nowadays excellent reviews over this topic exist (Rallison, 1984; Stone, 1994).

When an embedded drop is subjected to flow, the stresses of the external fluid on the drop surface are balanced by the interfacial tension. This stress field causes the deformation, orientation and possibly the break-up of the drop. In general, the response of the drop depends on: i) the viscosity ratio,  $\lambda = \mu_d / \mu_m$ , between the drop  $\mu_d$  and the fluid matrix  $\mu_m$ , as well as ii) the capillary number, defined as  $Ca = r_d \mu_m G / \sigma$ , where  $r_d$  is the equilibrium drop radius,  $G$  is the shear rate and  $\sigma$  is the interfacial tension. The capillary number ponders the ratio of effects due to viscous vs. interfacial stresses. Regarding the viscosity ratio, it is well established that there is a limit for the break up to occur when  $\lambda_{lim} \sim 3.6$  (Grace, 1982; Taylor, 1934); for values below this limit, the break up is possible if a critical Capillary number is reached or exceeded, otherwise, a steady deformation is reached. However, if the viscosity ratio is larger than  $\lambda_{lim}$  no breakups are observed and the drop will only attain a maximum deformation that depends on  $\lambda$ .

The quantification of droplet deformation is estimated with the deformation parameter  $D$ , initially defined by Taylor (Taylor, 1934) who performed the first important theoretical and experimental studies on drop deformation. For small deformations, the drop reaches an ellipsoidal shape (see Fig. 1). The deformation parameter,  $D$ , is defined in terms of the major and minor axis,  $L$  and  $B$  respectively, as

$$D = \frac{L - B}{L + B}. \quad (1)$$

Taylor derived the following analytic expression for the steady deformation  $D_0$ , in simple shear flow (Taylor, 1934),

$$D_T = Ca \frac{19 + 16\lambda}{19 + 19\lambda} \sin\theta \cos\theta, \quad (2)$$

where  $\theta$  is the orientation angle formed between the major axis and the principal flow direction.

However, Eq. (2) is only valid for  $\lambda < 1$  and  $Ca < 1$ . When  $\lambda \gg 1$ , Taylor obtained an asymptotic value for the deformation which is independent on  $Ca$  (Taylor, 1934),

$$D_T = \frac{5}{4\lambda}. \quad (3)$$

Despite advances of the previous investigations, there still are some important gaps in our understanding of the drop deformation dynamics. One of particular interest is the *transient response to start up flows* observed in high viscosity ratios. While in the case of low viscosity ratios, the temporal evolution towards a steady state of the parameters  $D_T$  and  $\theta$  are monotonic, for the case of high viscosity ratios,  $\lambda > \lambda_{lim}$ , the response is quite different and presents a

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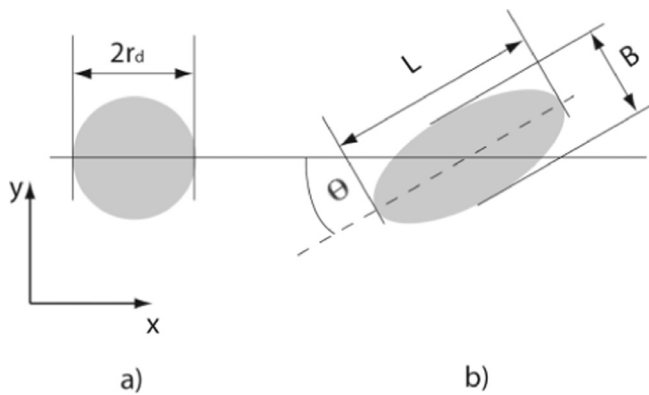


Fig. 1. Representation of an initial (a) and a deformed drop (b).

more complicated behavior (Cox, 1969; Rallison, 1980). Unlike systems with low viscosity ratio, the evolution to the steady state is no longer monotonic for high  $Ca$ , and stationary conditions are reached after a transient oscillatory behavior.

Experimentally this oscillatory behavior was first observed by Rumscheidt and Mason (1961) and Torza et al. (1972), who reported an oscillatory response in startup experiments in simple shear flow for high viscosity ratios. This transient behavior was also reported by Kennedy et al. (1994) in numerical simulations performed with high viscosity ratios, particularly for  $\lambda = 25$ , one of the experimental values used by Torza et al. (1972).

From a theoretical point of view, several models have been developed to describe the time dependent problem, among these, stand out those developed by Cox (1969) and Rallison (1980), based on the supposition that the deviation from initial sphericity of the drop is small and the instantaneous deformed drop is determined through a perturbation expansion in terms of spherical harmonics. For the case of high viscosity ratios with intermediate capillary numbers, both methods agree; however, Rallison's model is significantly more difficult to work with because it implies an equation that must be resolved numerically to obtain the time evolution of the drop deformation.

In clear contrast, the asymptotic model by Cox results in a simpler equation capable of describing the oscillations in the transient state of drop deformation and is valid for small deformations without restrictions on  $Ca$  or  $\lambda$ . The equation provided by Cox expresses the time evolution of the parameter  $D$  in simple shear flow as (Torza et al., 1972),

$$D(t) = D_0 \left[ 1 - 2e^{-\frac{20Gt}{19Ca\lambda}} \cos(Gt) + e^{-\frac{40Gt}{19Ca\lambda}} \right]^{1/2}, \quad (4)$$

Where  $D_0$  corresponds to the value of the steady state deformation given by:

$$D_0 = \frac{5(19\lambda + 16)}{4(1 + \lambda)\sqrt{(19\lambda)^2 + (20/Ca)^2}}. \quad (5)$$

Although this model reproduces the main feature of the transient state in high viscosity ratio (That is, the damped oscillations) and has a remarkably good accuracy about the damping rate, the coincidence with experiments and numerical simulations is merely qualitative since noticeable disagreement is observed, especially in the oscillation periods.

By observing Eq. (4), it is notorious that the oscillation periods are given by the shear rate applied. Low shear rates will produce extended periods and high shear rates will produce short periods. It is important to note that in non-dimensional time, the theoretical period for the oscillations is actually  $2\pi$ , a value that is constant not only for all capillary numbers but also for all viscosity ratios. This prediction of constant period is inaccurate since the first

experimental works from Torza et al. (1972), in which the data presented different oscillation periods for different values in the viscosity ratios. Besides, those experiments barely showed how the period behaves with different capillary numbers with a fixed viscosity ratio, whether it changes or not, as predicted by Eq. (4).

In this work, a modification to Cox model is developed; this modification allows a more accurate prediction of the oscillation periods by considering they depend on shear rate and the viscosity ratio. The modification was obtained from an analysis of the experimental and numerical data on quasi simple shear flow as well as from experimental data available in Torza et al., work (Torza et al., 1972).

## 2. Experimental part. Two-roll mills device

Experiments were carried out using the Two-Roll Mill Flow cell (Rosas et al., 2015). The flows generated by two-roll mills (2 RMs) device are in general non-linear, two-dimensional and capable of generating a wide spectrum of values of its kinematic parameters: in one side the flow is very close to simple shear, sweeping smoothly towards the other side where significant elongation effects can be present. An extensive description of the flows generated by 2 RMs device can be found in (Reyes, 2005; Reyes & Gefroy, 2000a, 2000b; Rosas, 2013). A detailed description of the flow cell geometry used along with the experimental procedure is available in (Rosas, 2013; Rosas et al., 2015). The flow field generated by the experimental device for this work corresponds to a flow type parameter  $\alpha = 0.03$  (Reyes, 2005) which because of the size of drops was very small compared to rolls gap, can be considered linear and indistinguishable from simple shear flow (Escalante-Velázquez et al., 2015).

The suspending fluid was PolyDiMethylSiloxane (PDMS), DMS T35 from Gelest Inc, with viscosity  $\mu_m = 51$  Poise and relative density  $\rho_m = 0.973$  at 25 °C. The drop fluid consisted of Polyisobutylene by Polysciences Inc., with a viscosity  $\mu_d = 800$  Poise and a relative density  $\rho_d = 0.92$  at 25 °C. For this system the viscosity ratio is  $\lambda = 16$  and the corresponding interfacial tension is  $\sigma = 3.5$  mN/m. All experiments were performed at 25 °C  $\pm$  0.1 °C.

## 3. Numerical method

Simulations of drop deformation between two cylinders, were carried out using the boundary element method (BEM). This method considers the boundary integral equations which can take into account the presence of rigid surfaces and deformable fluid interfaces (Escalante-Velázquez et al., 2015). The integral equation for the flow field within the drop,  $\mathbf{u}_2(\mathbf{x})$ , or exterior to the drop in the matrix fluid,  $\mathbf{u}_1(\mathbf{x})$ , will be dependent upon boundary integrals taken on the drop interface  $S_{\text{drop}}$  as well as on the cylinder surfaces  $S_{\text{rigid},1}$  and  $S_{\text{rigid},2}$ . These integral equations can be found in reference (Pozrikidis, 1992). In the present results, no wall effect was considering, so the ratio  $r_d/g$  (drop radius / cylinders gap)  $\ll 1$ .

In Fig. 2, a schematic representation is depicted. We assume a Newtonian, inertial-less drop with initial radius  $r_d$  and viscosity  $\mu_d$  —located between the two cylinders— and immersed in a Newtonian matrix fluid of viscosity  $\mu_m$ . The position of the stagnation point is located exactly halfway between the counter-rotating symmetric cylinders. The boundary of each cylinder and the drop boundary are discretized with 128 and 256 elements respectively. Given that previous studies have shown good agreement between numerical simulations and experimental deformations measured in high viscosity droplets (Escalante-Velázquez et al., 2015), two-dimensional (2D) numerical simulations were also considered for the present work.

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