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Effect of rheology of dense emulsions on the flow structure in agitated systems

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ABSTRACT

In manufacturing and applying emulsions it is often necessary to predict or control their flow and adjust their viscosity. A new method is presented for including the effect of the droplet size distribution on the rheological behaviour of dense oil-in-water emulsions of noncolloidal droplets. The method is based on extension of the advanced model for the relative viscosity of the concentrated monodisperse emulsion to account for polydispersity effects. The model includes effects of volume fraction of the dispersed phase, droplet size distribution, interfacial tension and shear rate on the relative emulsion viscosity. The model has universal character and can be combined with the population balance equation and CFD. Examples of modelling are presented for laminar and turbulent flows of dense emulsions. In the case of laminar flow the Couette flow and the Taylor–Couette flow are considered. In the case of turbulent flow it is shown how dispersion of droplets in the high-shear rotor-stator mixer affects the flow pattern and rheology of the emulsion product.

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1. Introduction

Emulsions are involved with many engineering operations and form many useful products. This includes emulsion-based food products, such as homogenized milk, sauces (mayonnaise, vinaigrette), dressings, beverages, butter, pharmaceutical products in the form of creams and balms. Emulsions are used in cosmetics, personal hygiene products, fire-fighting agents, agricultural industry products, paints and inks. In manufacturing and applying emulsions, it is often necessary to predict or control the flow of emulsions and manipulate the emulsion viscosity.

Emulsion viscosity η depends on the continuous phase viscosity η_c , the volume fraction of the dispersed phase ϕ and its viscosity η_d , the interfacial tension σ , the shear rate $\dot{\gamma}$, the emulsifying agent (if present) and of course on the temperature that obviously affects the physicochemical properties. Development of a strict, theoretical model is possible for infinitely dilute emulsions of spherical uncharged droplets that are very large compared to the continuous phase molecules, so that the Brownian motions can be neglected. The equation for the

relative viscosity η_r under limiting conditions when additionally the hydrodynamic viscous stress is negligible comparing to the interfacial stress was derived by Taylor (1932).

$$\eta_r = \frac{\eta}{\eta_c} = 1 + \left[\frac{5K + 2}{2(K + 1)} \right] \phi \quad (1)$$

where $K = \eta_d / \eta_c$ is the ratio of the dispersed-phase viscosity η_d to the continuous-phase viscosity η_c and ϕ is the volume fraction of droplets. Eq. (1) is valid for $\phi \rightarrow 0$, $N_{Ca} \rightarrow 0$ and $Pe \rightarrow \infty$, where N_{Ca} is the capillary number,

$$N_{Ca} = \frac{\eta_c \dot{\gamma} R}{\sigma} \quad (2)$$

being the ratio of the hydrodynamic stress that tends to stretch the droplet and increase its surface energy, to the interfacial stress resulting from interfacial tension, σ , tending to decrease the surface energy by maintaining its spherical shape. $\dot{\gamma}$ represents here the shear rate

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(the rate of deformation or extension) and Pe is the Péclet number that is defined as

$$Pe = \frac{\dot{\gamma}R^2}{D_B^\infty} \quad (3)$$

where D_B^∞ is the coefficient of Brownian diffusion for a particle separated from other particles, R is the drop radius, and $Pe \gg 1$ is equivalent to the condition that the drops are much larger than the distance travelled due to Brownian motions $R \gg (D_B^\infty/\dot{\gamma})^{1/2}$ during the characteristic deformation time, $\dot{\gamma}^{-1}$. For high values of the viscosity ratio, Eq. (1) reduces to the Einstein (1906) equation, valid for spherical, rigid particles at $\phi \rightarrow 0$.

$$\eta_r = 1 + 2.5\phi \quad (4)$$

Notice that in Eqs. (1) and (4) the hydrodynamic interactions between droplets and particles respectively are neglected due to large distance between them. Each drop just disturbs the flow of the continuous phase and the flow is generated within each drop independently of other drops, which affects the stress and the rate of energy dissipation.

In what follows we are interested in modelling viscosity of concentrated emulsions, for $\phi \gg 0$ and $N_{Ca} \geq 0$, which means that the interactions between droplets as well as droplet deformation from spherical shape and orientation of deformed droplets suspended in the continuous phase should be included. This is necessary, because as pointed out by Pal (2003a), rheology of emulsions is intimately linked to the deformation and orientation of droplets suspended in the continuous phase.

There are several equations for emulsion rheology in the literature, both theoretical and empirical, expressing the relative viscosity in terms of the volume fraction of the dispersed phase for diluted, concentrated and moderately concentrated emulsions including effects of the viscosity ratio and the capillary number. Also viscoelastic effects are included.

It is known (Pal, 1996) that the droplet size and the droplet size distribution have a significant influence on emulsion rheology. Fine emulsions have much higher viscosities than the coarse emulsions for the same volume fraction of the dispersed phase and the shear-thinning effect is stronger in the case of fine emulsions. Also the drop size distribution, i.e. proportion of coarse droplets to fine droplets, keeping total volume fraction of the dispersed phase constant, affects rheological properties. Salager et al. (2001) pointed out that the more polydispersed is the distribution, the less viscous is the emulsion, at all other characteristic parameters equal. This was observed when they compared the viscosity of the bimodal emulsion with the viscosity of the monomodal emulsion for the 70% oil content.

The aim of present work is to consider in the model effects of the drop size distribution on the viscosity, to describe some thixotropic effects as resulting from variation of the drop size population and link this model to CFD. A good start for this can be models by Pal (2003a,b, 2004). Pal employed the differential effective medium (DEM) approach to express the relative viscosity of the suspension of elastic particles, mimicking human erythrocytes (Pal, 2003b) and bubbly suspensions (Pal, 2004). For present paper the most important is the more universal model proposed by Pal (2003a) for concentrated emulsions using the analogy between shear modulus and shear viscosity, or more precisely using the form of Palieme (1990) model for shear modulus for dilute emulsions of noncolloidal droplets to express the shear viscosity.

The original model by Pal (2003a) was derived for mono-sized droplets, so in present work it will be extended to include effects of drop size distribution on emulsion rheology. This will be done using some elements of the original theory of Palieme (1990) that were not considered by Pal (2003a). To illustrate properties and application possibilities of the rheological model, it will be combined with the population balance, linked to CFD, and used to simulate the processes in both, laminar and turbulent regime. The model will be applied to simulate flow pattern of the laminar Couette and Taylor–Couette flows of dense emulsion. In the case of turbulent flow it will be shown how the drop dispersion and the highly turbulent flow in the high-shear rotor–stator

mixer affect rheological properties of emulsion and flow pattern during the process, as well as rheology of the resulting product emulsion.

2. Theoretical background and model presentation

Pal (2003a) presented in his paper three models to describe viscous behaviour of concentrated emulsions of two Newtonian, immiscible fluids, forming the monosize emulsion. All these models are based on the Palieme (1990) approach to modelling shear modulus, G . The most complex and most exact in Pal (2003a) paper is the model number 3. The model is constituted by Eq. (5) with three parameters M , N and P defined by Eqs. (6), (7) and (8) that depend on the capillary number $N_{Ca} = \eta_c \dot{\gamma} R / \sigma$ and the viscosity ratio $K = \eta_d / \eta_c$.

$$\eta_r \left[\frac{M - P + 32\eta_r}{M - P + 32} \right]^{N-1.25} \cdot \left[\frac{M + P - 32}{M + P - 32\eta_r} \right]^{N+1.25} = \left(1 - \frac{\phi}{\phi_m} \right)^{-2.5\phi_m} \quad (5)$$

$$M = \sqrt{(64/N_{Ca}^2) + 1225K^2 + 1232(K/N_{Ca})} \quad (6)$$

$$P = 8/N_{Ca} - 3K \quad (7)$$

$$N = \frac{(22/N_{Ca}) + 43.75K}{\sqrt{(64/N_{Ca}^2) + 1225K^2 + 1232(K/N_{Ca})}} \quad (8)$$

Using M , P and N from Eqs. (6), (7) and (8), one can calculate from Eq. (5) the relative viscosity, $\eta_r = \eta/\eta_c$, where η is the viscosity of emulsion, for any value of the capillary number N_{Ca} and viscosity ratio K . ϕ_m in Eq. (5) represents the maximum packing volume fraction. For $N_{Ca} \rightarrow 0$ Eq. (5) reduces to the Pal (2001) equation for concentrated emulsions $\eta_r [(2\eta_r + 5K)/(2 + 5K)]^{3/2} = (1 - \phi/\phi_m)^{-2.5\phi_m}$ that has been derived from Taylor (1932) equation (1) under assumption that the droplets are spherical at high concentration. For $N_{Ca} \rightarrow 0$ and $K \rightarrow \infty$ Eq. (5) reduces to the Krieger and Dougherty (1959) equation. The model expressed by Eqs. (5)–(8) was validated using many experimental data starting from the paper by Pal (2003a); this justifies its application in present work.

As mentioned earlier the Pal (2003a) model has been derived for mono-sized droplets. To use this model for polydisperse droplets we need to solve two problems. The first problem is related to the value of the maximum packing volume fraction, ϕ_m . In the case of monodisperse spherical droplets, so $N_{Ca} \rightarrow 0$, ϕ_m depends just on the type of droplet packing arrangement. It takes the value of 0.52 for cubic packing, 0.62 for random packing and 0.74 for hexagonal packing.

However, we are interested in polydisperse droplets, so the model should be modified. For polydisperse droplets with smaller droplets filling the space between the larger ones and due to drop stretching out into a filamentary shape, the value of ϕ_m can be much higher than 0.74. Howe and Clarke (1997) in the context of the Krieger–Dougherty (1959) model suggest $\phi_m = 0.8$ for polydisperse spheres. They argued that the exact choice of this parameter has relatively little effect on their results. Pal (2014a,b) for drops of non-uniform size applied the value $\phi_m = 0.85$. He applied the model of the type $\eta_r = f(\phi, \phi_m)$. Pal showed that using $\phi_m = 0.85$ the considered viscosity data of emulsions can be described accurately.

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