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Experimental investigation of viscoelastic drop deformation in Newtonian matrix at high capillary number under simple shear flow

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ABSTRACT

The steady state deformation of a viscoelastic drop (Boger fluid) in a Newtonian liquid at high capillary number under simple shear flow is investigated by direct visualization using a specially designed Couette apparatus which enables visualization from two perpendicular directions. Two drop deformation modes are found: (1) Mode I – drop deformation in the flow direction and (2) Mode II – drop deformation in the vorticity direction. The drop deformation mode depends on the relative strength of the elastic contribution to viscous contribution. If the elastic contribution is weak compared to the viscous contribution, the drop elongates in the flow direction via Mode I. If the elastic contribution is strong, the drop elongates in the vorticity direction via Mode II. The drop size also affects the drop deformation. At the same capillary number, bigger drops have larger deformations than smaller drops.

1. Introduction

Drop deformation in immiscible liquid blends presents itself in many chemical processes, such as cosmetics, paints, food and polymer processing. In these chemical processes, drop deformation determines the microstructure which in turn determines the macroscopic properties of the blends [1]. The deformation of a Newtonian drop in a Newtonian matrix has been extensively investigated by both experimental and numerical approaches providing a reasonably clear understanding on the essential physical mechanism [2–11]. However, drop deformation in viscoelastic fluid systems is much more complicated than that in pure Newtonian systems because of the complexity of the rheological properties and the modification of stresses and flow due to viscoelasticity [12–28].

To quantify drop deformation, Taylor [3] defined a deformation parameter *Df* in terms of the longest and shortest semi-axes of the drop cross-section (*L* and *B* respectively) as

$$Df = \frac{L-B}{L+B}. \tag{1}$$

The steady state drop deformation (*Df*) in pure Newtonian systems under simple shear with negligible buoyancy and inertial forces is governed by the viscosity ratio (η_r) of the drop phase and the matrix phase and capillary number (*Ca*) [3], i.e. *Df* is a function of η_r and *Ca*:

$$Df = f(\eta_r, Ca). \tag{2}$$

The capillary number is the ratio of shear stress to interfacial stress, which is defined as

$$Ca = \frac{\eta_m \dot{\gamma} R_0}{\Gamma}, \tag{3}$$

where η_m is the viscosity of the matrix phase, $\dot{\gamma}$ is the shear rate, R_0 is the initial drop radius and Γ is the interfacial tension between the drop and matrix phases. For a given Newtonian system, the drop deformation increases with increasing capillary number until the drop breaks up. It is relatively straightforward to investigate drop deformation in Newtonian systems. However, for immiscible liquid blends involving viscoelastic components, the study of drop deformation is more complex. The majority of viscoelastic materials, such as polymer melts, are usually elastic, highly viscous and shear-thinning, and therefore, very different drop deformation and breakup behavior is seen [29–33].

In order to obtain a clear understanding of drop deformation in viscoelastic systems, efforts have been made to separate the effect of elasticity from that of shear-thinning by using simple rheological models such as Oldroyd-B model for numerical studies [23,25] and by using model fluids such as Boger fluids for experimental work [17,19,34,35]. Similar to Newtonian systems, an analogous relation of drop deformation in viscoelastic systems should be written as

$$Df = f(\eta_r, Ca, \text{elastic properties}). \tag{4}$$

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The elastic properties are typically represented by two parameters: (a) Deborah number (*De*) which is a dimensionless number defined as the ratio of the material relaxation time (λ) to the characteristic time scale of an experiment (t_c, in shear flow, $t_c = 1/\dot{\gamma}$); or (b) the first normal stress difference (N_1). The viscoelasticity of drop phase has been demonstrated to inhibit drop deformation [13,36–39]. The critical capillary number for viscoelastic drop breakup is higher than that of its Newtonian counterpart [12,13,40]. The effects of elastic properties on drop deformation and breakup are often attributed to the first normal stress difference in viscoelastic fluids [14,17,36,38]. Aggarwal and Sarkar [25] developed a simple qualitative ordinary differential equation model on the force balance among shear stress, interfacial stress and the first normal stress difference to depict the essential physics underlying the experimental observations and numerical results. However, when the matrix is viscoelastic and drop is Newtonian, experimental results of the deformation contradict each other. Mighri et al. [13], Elmendorp and Maalcke [37] and Mechbal and Bousmina [39] found that matrix viscoelasticity facilitated drop deformation, while Flumerfelt [41], Guido et al. [19] and Verhulst and Moldenaers [42] found that matrix viscoelasticity resisted drop deformation and breakup. Yue et al. [23] clarified this non-monotonic behavior by investigating the modification of stresses and flow field by viscoelasticity.

Because of restrictions of experimental or computing capability, most previous work studied drop deformation in viscoelastic systems under simple shear over a limited range of small capillary numbers, typically below 1. At such low values of capillary numbers, the contribution of elasticity to the drop deformation is relatively small. Although Pillapakkam and Singh [18] simulated an Oldroyd-B drop sheared in a Newtonian matrix at high capillary number, the drop elasticity was still weak such that the drop broke up in the flow direction. However, in real industrial processes, the capillary numbers are usually very high. For example, the capillary numbers of polymer blending in extruders are on the order of 100. At such high capillary numbers, the drop deformation and breakup can be very different since elasticity is expected to dominate the deformation process at high shear rate. Hobbie and Migler [14], Migler [43] and Mighri and Huneault [17] found that at high capillary numbers with strong elastic effect, viscoelastic drops under shear elongated and broke up in the vorticity direction, which is the direction perpendicular to the flow direction and the velocity gradient direction. However, there is a lack of quantitative studies on drop deformation in viscoelastic systems sheared at high capillary numbers. Since the microstructure of many immiscible liquid blends is resultant from drop deformation and breakup at high capillary number, it is necessary to investigate the drop deformation under high capillary number.

In this work, we conduct a quantitative investigation on viscoelastic drop deformation in Newtonian matrix at high capillary number under simple shear flow, where the viscoelastic effects are strong and dominate the drop deformation process. We separate the effect of elasticity from that of shear thinning by using Boger fluids [44] as the drop phase.

2. Experimental

2.1. Materials

Table 1
Compositions of materials used in this study.

Designation		Material/formulation[a]	Viscosity, Pa s
Matrix	M1	PDMS(DMS-T31)	0.97
	M2	PDMS(DMS-T35)	4.86
	M3	PDMS(DMS-T41)	9.74
Drop	D1	0.6% PIB + 8.5% Ker + 90.9% PB	13.0
	D2	0.1% PIB + 4.9% Ker + 95.0% PB	14.5
	D3	0.8% PIB + 4.4% Ker + 94.8% PB	25.0
	D4	1.0% PIB + 5.8% Ker + 93.2% PB	31.0
	D5	1.3% PIB + 7.2% Ker + 91.5% PB	40.0

[a] All percentages are on mass basis.

Three low-viscosity polydimethylsiloxane (PDMS, Gelest, Inc.) were used as matrix phase fluids (see Table 1). PDMS liquids with such low viscosities exhibited Newtonian properties under the experimental conditions studied. The drop phase fluids were polymer solutions with shear-independent viscosity, also known as Boger fluids, which were prepared under slow mixing for a period of more than 5 days to ensure molecular level dissolution of the polymers. The Boger fluids used in this study were mixtures of polyisobutylene (PIB, Mw = 1×10^6 kg/kmol, Sigma-Aldrich), kerosene (Ker, Fisher Scientific), and polybutene (PB, Mn = 920 kg/kmol, Sigma-Aldrich). The properties and formulations of the materials are listed in Table 1. The interfacial tension between the Boger fluid and PDMS is 3 mN/m [17]. Results from Guido et al. indicate that the miscibility between the drop and matrix phases is not an issue to study the drop deformation and breakup [19].

Fig. 1 shows the viscosity (η) and the first normal stress difference (N_1) of the Boger fluids, which were obtained using a Rheometrics RMS800 rheometer operated in steady mode with

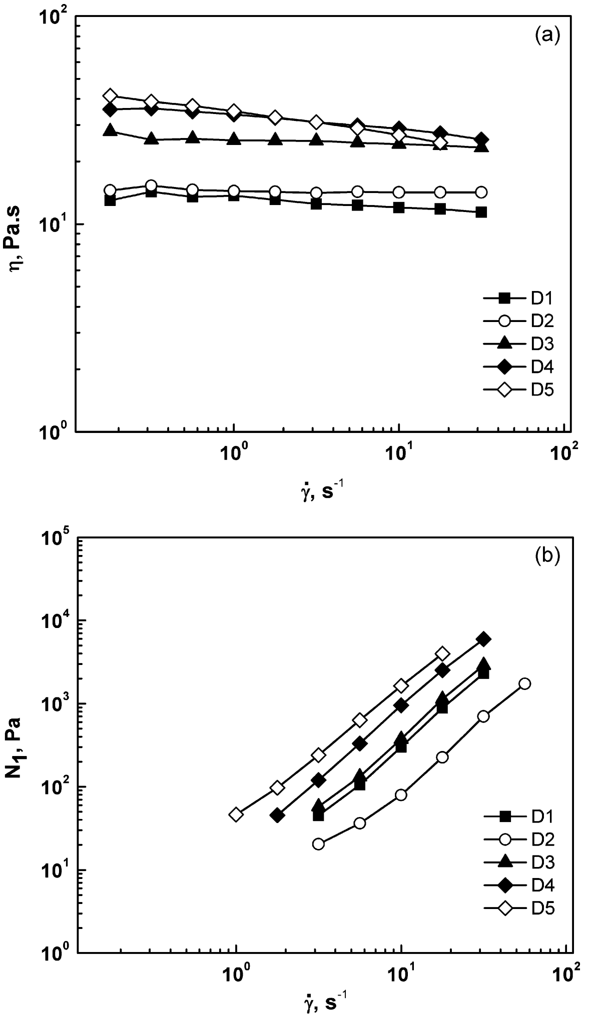

Fig. 1. Rheological properties of Boger fluid. (a) Viscosity η. (b) The first normal stress difference N_1.