

## CHAPTER II

### LITERATURE SURVEY

The deformation and breakup of a suspended immiscible Newtonian droplet in another Newtonian matrix in a steady state shear flow was first pioneered and studied by Taylor (1932, 1934). He suggested that the droplet deformation is controlled by two dimensionless numbers: the viscosity ratio,  $\eta_r$  which is the ratio between viscosity of droplet (disperse phase) ( $\eta_d$ ) and viscosity of matrix ( $\eta_m$ ), and the Capillary number ( $Ca$ ) or Taylor number, defined by the viscous force to the interfacial surface force:

$$Ca = \frac{\gamma \eta_m D_0}{2\Gamma} \quad (2.1)$$

where  $\gamma$  is the shear rate,  $D_0$  is initial droplet diameter and  $\Gamma$  is interfacial tension. For small deformation, the shape of droplet gradually becomes ellipsoidal while its deformation, written as a function of its lengths, is given by:

$$Def \equiv \frac{a-b}{a+b} = Ca \frac{19\eta_r + 16}{16\eta_r + 16} \quad (2.2)$$

where  $a$  and  $b$  are the lengths of the major and minor axes of the deformed droplet (ellipsoidal shape), respectively. Taylor predicted that the critical point at which the viscous force overcomes the interfacial force leading to droplet breakup occurs at  $Ca_c$  (critical capillary number)  $\approx 0.5$  and  $Def_c$  (critical deformation)  $\approx 0.5$  for a steady simple shearing flow (or quasi-steady), if the flow rate is very slowly increased with a viscosity ratio of around unity. Here, the word critical means the condition at breakup. These basic predictions have been confirmed by many experiments (Rumscheid and Mason 1961; Grace 1982; Bentley and Leal 1986; Guido and Villone 1997). When the viscosity ratio is higher than four, no breakup occurs (Grace, (1982)). These results show that for Newtonian fluids, droplet deformation and breakup is strongly influenced by viscosity ratio. Grace (1982) found that, for steady state shearing of an isolated

Newtonian droplet in a Newtonian matrix, the minimum critical capillary number occurs when the viscosity ratio is around unity.

The breakup of isolated viscoelastic drops in a Newtonian matrix and of Newtonian drops in a viscoelastic matrix in simple shear flow was investigated by Elmendorp and Maalcke (1985) with particular emphasis on the effect of elasticity. Their results showed that the droplets, which have higher elasticity, have more ability to resist against breakup because they tend to stabilize themselves. In contrast, the deformation of a Newtonian drop in a viscoelastic matrix increases with increasing matrix elasticity. De Bruijn (1989) reported that the critical capillary number for viscoelastic drops sheared in a Newtonian matrix is slightly higher than the critical capillary number for Newtonian drops sheared in a Newtonian matrix. He found that the droplets break most easily when viscosity ratio is between 0.1 and 1. On the other hand, the droplets do not break when the viscosity ratio is higher than four. This result confirms the results reported by Grace (1982).

Mighri *et al.* (1998) investigated the influence of elastic properties on drop deformation and breakup in shear flow. They defined the elasticity ratio,  $k'$ , as ratio of characteristic elastic time ( $\lambda$ ) of the droplet phase to the matrix phase, where  $\lambda = N_{11}/2\eta\dot{\gamma}^2$  and  $N_{11}$  is the first normal stress difference. Their results show different mechanisms of the drop deformation between the elastic system and the Newtonian system of approximately the same viscosity ratio and the same interfacial tension. The steady-state drop deformation for shear rate less than critical value is affected by both the drop and matrix elasticities. The matrix elasticity helps to deform the drop, whereas the drop elasticity resists the drop deformation. For high elasticity ( $k' < 0.37$ ) the deformation of elastic drops in an elastic matrix under shear is higher than that of Newtonian drops in a Newtonian matrix. However, for  $k' > 0.37$ , the elastic drops deform less than Newtonian drops in a Newtonian matrix. Thus, the droplet resistance to deformation and breakup increases with increasing elasticity ratio of droplet to matrix phase.

The alignment of droplet under shear flow was observed by Migler (2000). The deformation of elastic droplets in a polymeric matrix under a shear flow was studied in order to find the conditions in which droplets can align in the vorticity direction. The viscosity ratio is near unity, but the elasticity ratio of the droplet to the matrix is greater than 100. Under this condition, the matrix phase was nearly Newtonian. In the limit of weak shear and small droplets ( $Ca < 5$ ), the droplet alignment was found to be along the flow direction (shear direction), in contrast for a strong shear and large droplets ( $Ca > 5$ ), the droplet alignment was along the vorticity direction with a broad distribution of aspect ratios.

Hayashi *et al.* (2000) observed the shape recovery of a deformed droplet in an immiscible polymer matrix under large step strains. They discovered that after applying a large step strain, a spherical droplet in a matrix with higher viscosity deformed to a flat ellipsoid. Then the flat ellipsoid changed into a rod-like shape, to a dumbbell and to an ellipsoid in revolution. Finally, its shape relaxed back to the sphere. The shape recovery can divide into three stages. The early stage of the shape recovery, the large reduction of surface area occurs by changing the shape from the flat ellipsoid to the rod-like without reducing droplet stretch. At the intermediate stage, the further reduction occurs slowly by reducing the droplet stretch. The capillary instability also appears at this stage but the reduction of the surface area due to this instability is small. In the last stage, the droplet recovers the spherical shape depending on the initial radius of droplet. The time needed for this process increases with the increasing initial radius of droplet and with increasing strain. They also observed the orientation angle between the major axis and the shear direction (flow direction) during this recovery process. They found that the orientation remained the same.

Mighri and Huneault (2001) studied the deformation and the breakup of a single droplet of a viscoelastic Boger fluid in a Newtonian matrix, sheared in a transparent Couette flow cell. At low shear rate, they found that the steady-state deformation increased with shear rate as expected, but above a critical shear rate ( $Ca \approx 5$ ) the deformed drop began contracting in the flow direction and changed its orientation

to the vorticity axis. With further increases in shear rate, this elongation in the vorticity direction increased until breakup finally occurred at a capillary number no higher than  $Ca \approx 35$ . They proposed that the critical shear stress for reorientation of the droplet in the vorticity direction was probably related to the values of the first and second normal stress differences and their dependencies on shear rate.

Newtonian drops in Newtonian suspending fluid and Newtonian drops in viscoelastic suspended fluid were investigated by Ha and Leal (2001). The stability of drops following the cessation of flow in each case was determined. If the ratio between extended lengths to initial radius is high enough, the drops will break into two or more parts as known as end-pinching. However, in the Newtonian/Newtonian system, the critical degree of stretch for breakup increases with increasing capillary number. Moreover, for a Newtonian drop in a Newtonian fluid, their stability is slightly less than that of the same drops in a viscoelastic fluid.

Lerdwijitjarud *et al.* (2003) showed that in a 20% blend of a Newtonian liquid in a Newtonian matrix the steady-state capillary number is actually lower than the critical capillary number for breakup of an isolated droplet, i.e., the disturbances to the flow produced by the presence of other droplets enhance breakup of a given droplet to an extent that more than offset any increase in droplet size due to coalescence. Thus, the high steady-state capillary numbers observed in blends of viscoelastic polymers must be attributed to the role of viscoelasticity.

Recently, single viscoelastic droplets in Newtonian or viscoelastic matrices have been observed microscopically in simple shearing flows. Lerdwijitjarud *et al.* (2003) observed deformation and breakup of isolated droplets of weakly elastic fluid ( $Wi_d \leq 0.02$ ) in a Newtonian matrix, and found that droplet elasticity produces a slight (up to around 20%) increase in  $Ca_c$ , the critical capillary number for droplet breakup. The breakup mechanism appeared to be similar to that in a Newtonian fluid; i.e., the droplet deformed increasingly in the flow direction as the shear rate was gradually increased, until breakup occurred. Elasticity of the droplet produced a reduction in the degree of deformation at any given shear rate and a greater critical deformation at

breakup, resulting in a higher  $Ca_c$ . However, at the highest Weissenberg number, this effect appears to be saturating, leading to only a modest increase in  $Ca_c$ .

In 2004, Lerdwijitjarud *et al.* studied the elasticity of droplet by adding high molecular weight polybutadiene into low molecular weight polybutadiene dispersed phase and blend with polydimethylsiloxane matrix. They found that the steady state deformation of isolated droplet decreased with increasing dispersed phase elasticity at the same imposed capillary number. They also found the relationship between critical capillary number ( $Ca_c$ ) and dispersed phase Weissenberg number ( $Wi_d$ ). The linear relationship holds up to a value of  $Wi_d$  equal to one and  $Ca_c$  approach to asymptotic value of 0.95 for high  $Wi_d$ . The steady state capillary number ( $Ca_{ss}$ ) that calculated from average droplet diameter for 10%-dispersed phase blends is less than the  $Ca_c$  for isolated droplets for the same blends.

Cherdhirankorn *et al.* (2004) measured the dynamics of deformation of an elastic droplet in an elastic matrix. Both two polymers have the same viscosity but differing degree of elasticity. The different elasticities produce qualitative differences in the droplet deformation occurring before the droplet attains its steady state shape. For the system with higher elasticity, the deformation oscillates several times before reaching its steady state shape, but for the lower elasticity system, the droplet deforms in shear direction first and continuously contacts in the flow direction until reaching its steady state shape. The capillary number can be varied at fixed elasticity and shear rate by increasing the droplet size. The steady state shape of both systems deform increasingly along the vorticity direction.