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MIXING OF IMMISCIBLE LIQUIDS

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1. INTRODUCTION

The 'dispersive mixing' of immiscible liquids is an essential subject in many industrial processes involving multiphase flow. For example, in the melt blending of incompatible polymers the morphology of the blend is of major influence on its mechanical (and other) properties. Therefore, it is important to model the development of morphologies during the mixing process; starting with large (mm) domains and ending with a distribution of small (μm) domains of the dispersed phase (ref. 1).

The deformation of a dispersed liquid domain in a flow field of the continuous phase is governed by the Capillary number, defined as the ratio of the (deforming) shear stress τ and the (conservative) interfacial tension σ scaled on the characteristic radius of curvature of the interface:

$$Ca = \frac{\tau}{\sigma/R} \qquad (1)$$

During the initial stages of mixing the radius R is large (typically order of mm) so that τ is dominant over σ/R. In this case, only distributive mixing occurs and the parameters of interest are the total deformation applied by the compounding equipment and the number of folds or reorientations. The dispersed drops deform affinely with the flow field of the matrix.

Once thin liquid threads have been formed by affine deformation of the dispersed phase, local radii have decreased such (μm) that the interfacial stress σ/R becomes important. Distortions at the interface grow and cause breakup of the threads into lines of small droplets (dispersive mixing). These droplets may deform and break up again in the flow field or they may resist further deformation if they are small enough (σ/R dominant over τ). In other words: Drops do not break up if Ca is below a certain critical value Ca_{crit}. This value is a function of the type of flow and the viscosity ratio p between dispersed and continuous phase:

$$p = \eta_d / \eta_c \qquad (2)$$

Apart from breakup of drops via threads into smaller drops also coalescence of colliding droplets may occur, coarsening the morphology.

A classical way to model the mixing process of immiscible liquids is to study the deformation and breakup of a single dispersed drop using model liquids (at roomtemperature) in well defined flows. This study presents some results on distributive ($Ca >> Ca_{crit}$) and dispersive ($Ca \approx Ca_{crit}$) mixing in plane elongational flow, generated in an opposed jets device. Special attention is paid to viscoelasticity of the dispersed phase.

2. THE OPPOSED JETS DEVICE

In studies on drop deformation and breakup, a variety of experimental devices is used to generate specific types of flow (e.g., a Couette device for simple shear flow or a four roll mill for plane hyperbolic flow). For the present study a so called opposed jets device was developed to generate plane hyperbolic flow (ref. 2). The principle is shown in Fig. 1 and is based on the stagnation flow of two opposed jets that are forced inbetween four solid blocks, fixed between two transparant parallel plates. Advantages over the four roll mill are the small size of the device and its simplicity. Numerical simulations have shown that in the central region the flow field is plane hyperbolic (2D elongational), obeying the velocity field:

$$u = Gx, \qquad v = -Gy, \qquad w = 0 \tag{3}$$

where G (s^{-1}) is the velocity gradient or rate of elongation. From the simulations it turned out that G is more uniform (as it should be) in an opposed jets device than in a four roll mill. Experimental verification of the velocity field, using laser Doppler anemometry, confirmed the central area of constant G.

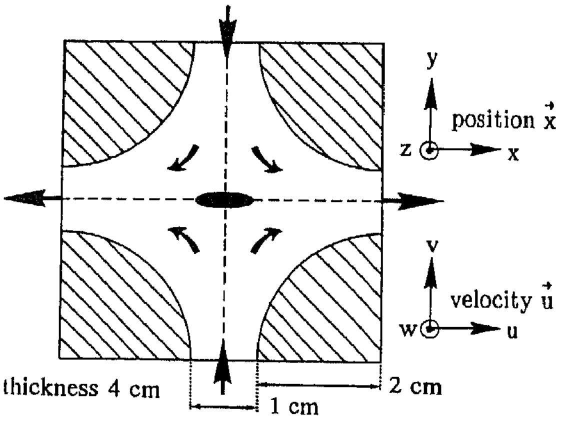

Fig. 1 Principle of the opposed jets device for drop deformation in plane hyperbolic flow.

An automatic control system is required to keep the deforming drop at its principally unstable position in the stagnation point, which it tends to leave. A control cycle consists of determination of the drop position (using a camera and an image processor), calculation of the required flow adjustment, and realization of the control action. Flow adjustment is carried out by decreasing the exit flow rate in the direction of the drop movement. In this way, the stagnation point is transferred beyond the drop forcing it to return to the centre. A sample rate of 25 control cycles per second results in a stable position of the deforming drop at the centre of the device at flow rates up to G ≈ 10 s^{-1}. Deformation of a dispersed (viscous or viscoelastic) drop or thread in elongational flow can now be studied using videorecordings through a microscope.

3. RESULTS ON DISTRIBUTIVE MIXING

If the applied Capillarynumber amply exceeds the critical value necessary for drop breakup, i.e. if $Ca >> Ca_{crit}$, the drop is expected to deform affinely with the global flow field without any significant resistance of the interfacial tension. In practice, this is the case for large drops ($Ca \sim R$), thus in the initial stages of mixing. Using the opposed jets device, this can be simulated as follows.

A 1 mm drop of castor oil (Newtonian, viscosity 0.74 Pa·s) is subjected instantaneously to a steady plane hyperbolic flow of silicon oil (Newtonian, viscosity 0.94 Pa·s, interfacial tension to castor oil $4.1 \cdot 10^{-3}$ N/m). The velocity gradient G is made such that Ca exceeds Ca_{crit} with different ratios. In plane hyperbolic flow (ref. 1):

$$Ca = \eta_c 2GR / \sigma \tag{4}$$

A measure for the deformation of the drop, with long and short axes L and B, is usually chosen as:

$$D = (L-B) / (L+B) \tag{5}$$

and ranges from 0 for a sphere to 1 for an infinitely long and thin body. Upon integrating Eq. (3), it can be seen that if the drop deforms affinely with the matrix, it elongates exponentially with time (t); D is a function only of the applied deformation Gt:

$$D = (\exp(2Gt)-1) / (\exp(2Gt)+1) \tag{6}$$

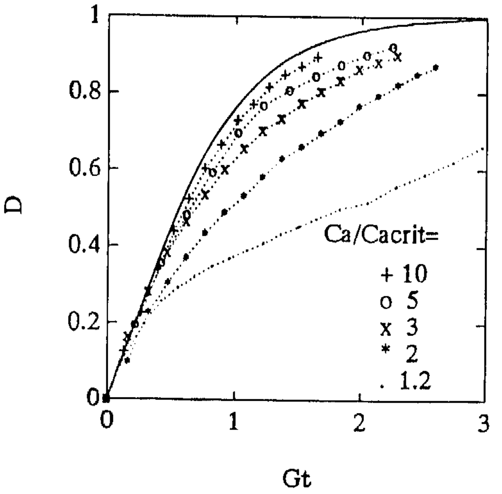

Fig. 2 Deformation of an initially spherical drop in a uniform plane hyperbolic flow at different ratios of exceeding Ca_{crit}; the full curve corresponds to affine deformation, Eq. (6).

In Fig. 2 the measured progress of D is shown. It can be seen that on exceeding Ca_{crit} the deformation tends to the theoretical curve for affine deformation. Using conservation of volume the width of the drop in z-direction can be estimated from L and B (measured directly). It seems that as the deformation progresses affinely this third drop axis remains constant (as it should, since w = 0). So the 3D shape of the drop in plane hyperbolic flow is like an ellipsoid with non-circular cross section.

4. RESULTS ON DISPERSIVE MIXING

As the dispersed drops have been elongated into long thin liquid threads, these threads become unstable. Very small distortions that are always present at the interface between thread and continuous phase are favoured to grow due to the interfacial tension. Depending on the viscosity ratio, distortions having one specific wavelength grow fastest and cause breakup of the thread into a line of small droplets. For the case of Newtonian liquids and without any external flow field the exponential growth of these sinusoidal 'Rayleigh distortions' can be calculated (ref. 3). An external flow field complicates the problem since the thread radius continuously decreases and the actual fastest growing wavelength is stretched so that it is no longer dominant: a new wavelength has to develop. As a consequence, breakup times of threads are increased by an external flow field (ref. 4).

The effect of viscoelasticity of the thread phase on the breakup of threads is even more dramatically. Initially, distortions grow even faster than in Newtonian systems (ref. 5, 6). But at a certain amount of deformation the amplitude almost stops growing and dumbbell shaped threads remain for a relatively long time (Fig. 3). This is due to built up elongational stresses in the connecting fibers, drastically increasing the elongational viscosity and thus inhibiting further drainage from the fibers into the bulbs. Breakup times of viscoelastic threads can be orders of magnitude longer than comparable (i.e. viscosity) Newtonian threads. Once a fiber has broken, the remaining parts elastically retract to neighbouring drops.

The influence of viscoelasticity of the dispersed phase on the value of Ca_{crit} (necessary for drop breakup) is much less. From experiments in the opposed jets device it appears that for viscosity ratios around unity Ca_{crit} does not significantly deviate from Newtonian systems (also ref. 6, 7).

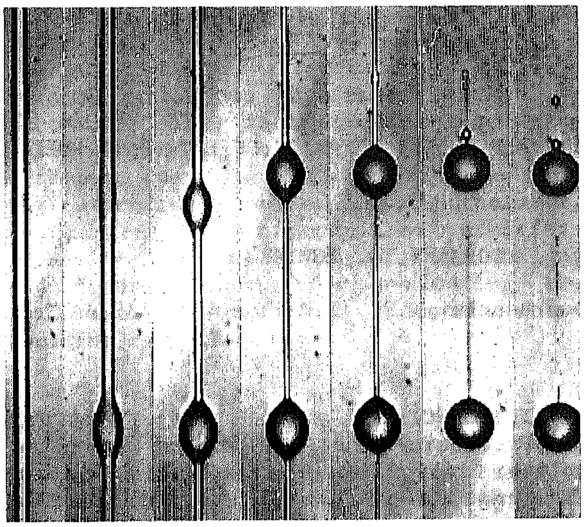

Fig. 3 Disintegration of a 0.07 mm viscoelastic thread (80% corn syrup/ 20% water/ 0.01% polyacrylamide) in a quiescent Newtonian matrix (silicon oil). Characteristic is the dumbbell shape. Photographs were taken after every 3 seconds.

5. NUMERICAL APPROACH

Progress is made in the numerical simulation of the drop / thread deformation problem. A finite element approach was chosen with an 'arbitrary Lagrange Euler' (ALE) mesh formulation. First the Newtonian problem has been solved whereafter a viscoelastic element is incorporated.

6. CONCLUSIONS

The opposed jets device is well suitable for drop deformation experiments. Upon sufficient excess of Ca_{crit}, drops deform nearly affinely. Viscoelasticity of the dispersed phase strongly decelerates the final stage of thread breakup.

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