EFFICIENT HOMOGENISATION OF PHOTOGRAPHIC DISPERSIONS

The formation of fine droplets in a photographic emulsion which is forced through an orifice disperser consisting of a tube with one or more abrupt constrictions is considered. Some design ideas for reducing the droplet size are presented.

1. Introduction

During the manufacture of colour photographic paper, the paper surface is overlaid with expensive dye-forming chemicals. These chemicals are dissolved in oil and are applied to the paper surface as an emulsion of small droplets in an aqueous phase. It is desirable that the droplets be made as small as possible. Smaller droplets would result in higher quality photographs for a given quantity of chemicals. Alternatively, smaller droplets would achieve a cost saving for a given photographic quality, by requiring a smaller quantity of chemicals.

The final homogenisation of the emulsification is achieved by forcing a relatively coarse oil-aqueous mixture containing 1–2 μm drops through an orifice disperser (Figure 1) to create droplets with a mean diameter of about 0.1–0.2 μm. The tube diameter $D = 5$ mm, and the orifice typically has diameter $d_o = 1$ mm, giving an area contraction ratio of 25:1. Strength requirements for the disperser dictate that the orifice length must be at least 3 mm. The available pressure drop is limited to 50 MPa.

The MISG problem, presented by Kodak (Australasia) Pty Ltd, was to identify changes to the disperser geometry, subject to a fixed pressure "budget", which would result in a lower mean drop size. Because of the cost of the photographic chemicals and the quantity used world wide by Kodak, even a small reduction in the final mean drop diameter from, say, 0.11 to 0.10 μm would be significant.

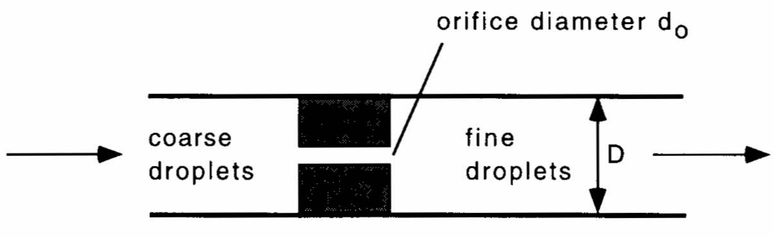

Figure 1: Schematic of the Kodak orifice disperser.

2. Notation

D	–	tube diameter
d_o	–	orifice diameter
Q	–	volumetric flow rate
ΔP	–	pressure drop
C_D	–	discharge coefficient
A_o	–	orifice area
ρ	–	mixture density
Re_o	–	orifice Reynolds number for the mixture
Ca	–	Capillary number
G	–	shear rate
η	–	bulk viscosity
η_C	–	viscosity of continuous (aqueous) phase
η_D	–	viscosity of droplet phase
γ	–	interfacial tension
r	–	droplet radius
$\overline{U_o}$	–	mean velocity in the orifice
t	–	time
t_{def}	–	time scale for drop deformation
θ	–	ratio of final to initial drop radius
R	–	orifice radius ($\frac{1}{2}d_o$)
y	–	boundary layer thickness
x	–	axial distance
σ	–	Cavitation number
P_v	–	vapour pressure

3. Material properties

The photographic chemicals are dissolved in the oil phase. The aqueous (gel) phase consists of water with about 15 per cent (by volume) of dissolved gelatine together with about 0.5 per cent surfactant. The oil-aqueous mixture (emulsion) which is passed through the disperser contains 10–20 per cent by volume of oil. The temperature of the flow is given as 80°C. Table 1 gives the viscosity and density range of each phase at this temperature. The deliberations of the MISG were based on the specific typical values also shown in Table 1. Experiments performed by Kodak showed that the mixture is only slightly shear thinning and that the viscosity is almost constant for shear rates greater than 10^3 s^{-1}. Calculations by the finite element numerical package *Fastflo* (Luo *et al.*, 1996) indicated that the shear rates in the orifice are typically orders of magnitude greater than this value. We therefore concluded, with some relief, that the mixture is approximately Newtonian for the shear rates obtained in the disperser.

viscosity (aqueous)	5–20 cP	(15 cP)
viscosity (oil)	10–500 cP	(50 cP)
viscosity (bulk)		(40 cP)
specific gravity (aqueous)	1.0–1.2	(1.0)
specific gravity (oil)	0.9–1.2	(1.0)
Interfacial tension	3–20 dyne/cm	(10 dyne/cm)

Table 1: Ranges of physical properties. Typical values used by the MISG are bracketed.

4. Orifice flow

The flow rate (Q) through the orifice depends on the applied pressure drop (ΔP) across the orifice and the flow resistance which is usually expressed by an empirical discharge coefficient C_D. For a narrow, sharp-edged orifice having a small orifice area (A_o) compared with the area of the pipe (Coulson and Richardson, 1990; p. 210)

$$Q = C_D A_o \sqrt{2\Delta P/\rho} \tag{1}$$

where ρ denotes the fluid density. For orifice Reynolds numbers ($Re_o = \rho \overline{U_o} d_o/\eta$) exceeding 1000, C_D lies in the range 0.6–0.65. Appropriate C_D values are not known for the Kodak disperser with its much longer orifice (at least 3 mm), but we expect them to be smaller because of increased frictional resistance.

A consequence of equation (1) is that the fluid velocity in the orifice ($\overline{U_o} = Q/A_o$) is independent of the orifice diameter d_o (assuming a constant value of C_D), and hence Re_o is linear in d_o.

For pressure drops ranging between 10 and 50 MPa, equation (1) predicts flow rates in the range 4–10 l/min, and Re_o in the range 2000–5000, for a single 1 mm diameter orifice and a bulk viscosity of 40 cP. The corresponding inlet Reynolds number range is 400–1000. Obviously, smaller pressure drops will result in lower values of flow rate and Reynolds number. If the orifice in the Kodak disperser offers more flow resistance than this example case, as discussed above, then Q and Re will again be lower. This is consistent with the example flow rate of 3 l/min quoted by Kodak.

The mixture emerges from the orifice as a high speed jet ($\sim$ 100 m/s) surrounded by one or more toroidal recirculation cells. Nevertheless the Reynolds numbers in the orifice and upstream are such that the flow is in transition between laminar and turbulent flow, with the flow becoming fully turbulent downstream of the orifice. It is possible that pressures in the recirculation zone become sufficiently low that cavitation occurs. The axial extent of the recirculation zone is defined by the jet reattachment length which is expected to be about 8 tube diameters for this contraction/expansion ratio (Agarwal, 1994). Downstream of the jet reattachment, pressure recovery would result in the collapse of cavitation bubbles. A schematic of the expected flow patterns is shown in Figure 2.

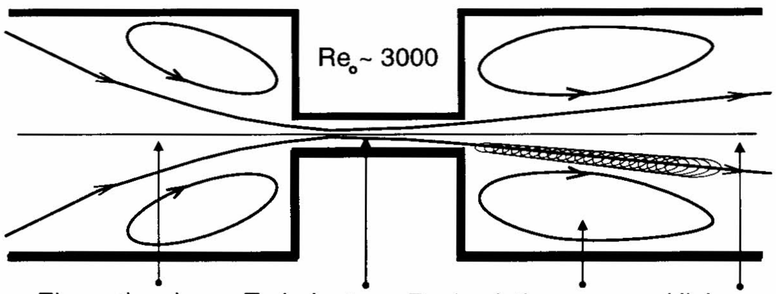

Figure 2: Schematic of likely flow patterns in the orifice disperser.

Thus, the mechanisms which can deform and subsequently break droplets in the Kodak disperser include (i) laminar elongation and shear in the flow entering the orifice, (ii) turbulent stresses in the developing jet, and (iii) turbulence and (possibly) cavitation downstream of the recirculation zone.

5. Droplet breakup

Walstra (1983) gave an excellent review of the physics of droplet formation in emulsions, in viscous (laminar), turbulent, and cavitating flows. More recently, Stone (1994) reviewed the dynamics of droplet breakup in viscous flow. We consider, in turn, the implications of the following for droplet breakup: (i) the laminar flow at the orifice entry, (ii) the growth of the turbulent boundary layer within the orifice, and (iii) possible cavitation in the flow exiting the orifice.

5.1 Orifice entry flow

Drops in the Kodak disperser experience a time-dependent shear history, whereas published experiments and theory which best describe drop deformation relate to drops in steady shear. Thus only rough estimates of conditions for drop breakup can be derived.

A drop of radius r will break when the capillary number

$$Ca = \frac{G \eta_C r}{\gamma} \tag{2}$$

exceeds a critical value Ca_{crit}. The capillary number represents the ratio of viscous stresses ($\eta_C G$) to the Laplace pressure difference (γ / r) which resists drop deformation.

The critical capillary number depends on the flow type (e.g. simple shear versus elongational flow) and on the viscosity ratio η_D / η_C of the dispersed (oil) to continuous (aqueous) phase. The converging flow entering the orifice is necessarily elongational (i.e. there is a velocity gradient in the direction of flow) and, typically, $\eta_D / \eta_C \sim 3$. In that case, $Ca_{crit} \sim 0.1 - 0.2$ (Stone, 1994). Thus we expect that the rate of strain G must satisfy

$$G > G_{crit} \sim 10^6 \text{ s}^{-1} \tag{3}$$

for drops larger than 0.1 μm to break.

In elongational flow with viscosity ratios present in the Kodak disperser, the drop is pulled into a thinning thread before breaking into droplets. Fragmentation can be triggered by capillary instabilities on very long threads, and "end-pinching" during transient stretching and relaxation (Stone, 1994). Final droplet size depends on the thread diameter at breakup. Higher viscosity drops will result in thinner threads before breakup and hence smaller droplets.

Using $G \sim \overline{U_o} / d_o$ gives a G value of the order of G_{crit} for pressure drops in the range 10–50 MPa when $d_o = 1$ mm, the orifice diameter currently used by Kodak. Similarly, *Fastflo* laminar flow calculations for $Re_o = 2000$ also predict $G \sim G_{crit}$. (That calculation is not strictly valid since flow is in transition, and the results are not considered completely reliable due to convergence difficulties). Since the breakup of drops down to about 0.1 μm diameter is known to occur, the above estimates of G near the critical value in equation (3) suggests that the orifice entry is responsible for the breakup of drops to some intermediate size, and that factors such as turbulence and cavitation complete the final size reduction.

A further requirement for the breakup of a drop is that it must experience this rate of strain for a time $t > t_{def} = \eta_D r / \gamma$ where t_{def} is the time scale for drop deformation. It is expected that the time needed for breakup is several times that needed for deformation. For breakup of drops with diameters exceeding 0.1 μm, we therefore require

$$t > t_{def} \sim 10^{-6} \text{ s.} \tag{4}$$

We expect this condition to be well satisfied since a time scale for the flow based on axial velocity and orifice diameter lies in the range 10^{-5}–10^{-4} s.

An optimal d_o?

From Walstra (1983; p 83), and since $Ca \propto G$,

$$\theta \propto G^{-\frac{2}{3}} \tag{5}$$

where θ denotes the ratio of final to initial drop radius based on the breakup of extending liquid threads in elongational flow. If the flow entering the orifice (radius $R = \frac{1}{2}d_o$) is modelled as a point sink then

$$G \propto QR^{-3}. \tag{6}$$

Two limiting flow profiles within the orifice are considered: (i) plug flow to represent the relatively flat velocity profile in fully developed turbulent pipe flow, and (ii) Poiseuille flow corresponding to fully developed laminar flow in a pipe. For plug flow, $Q \propto R^2$, and for Poiseuille flow with a given pressure gradient, $Q \propto R^4$. Equations (5, 6) then give

$$\theta \propto R^{\frac{2}{3}} \quad \text{for plug (turbulent) flow} \tag{7}$$

$$\theta \propto R^{-\frac{2}{3}} \quad \text{for Poiseuille (laminar) flow.} \tag{8}$$

Thus, reducing R is predicted to yield smaller droplets in plug flow, whereas increasing R is predicted to do so in Poiseuille flow. For both cases Re_o increases with R, hence the flow is laminar when R is small and turbulent when R is large. Thus, θ first decreases (equation (8)) and subsequently increases (equation (7)) with increasing orifice radius. This implies that there exists an orifice diameter which is optimal for droplet breakup.

5.2 Turbulent boundary layer

Within the orifice ($Re_o \sim$ 2000–5000), it is expected that a turbulent boundary layer grows from the entry corner. Its growth on a wall is given by (Davies, 1972; p. 45)

$$\frac{y}{x} = 0.376 Re_x^{-0.2} \tag{9}$$

where x denotes the axial distance from the orifice entry and y denotes the thickness of the boundary layer. For the orifice, the Reynolds number Re_x is approximated as $\rho\overline{U_o}x/\eta$. Equation (9) then yields $y/x \sim 0.05$ in which case the entire orifice cross-section would become turbulent if the orifice was extended to 10–20 orifice diameters in length. Since turbulence is one of the mechanisms for drop breakup, it may be desirable to lengthen the orifice from the minimum 3 mm permitted to, say, 10 mm for a 1mm diameter bore.

5.3 Downstream cavitation

Along with turbulence in the downstream flow, the possibility of cavitation exists in the low pressure recirculation adjacent to the jet exiting the orifice. It occurs when the pressure falls below the vapour pressure of the liquid. Cavitation bubbles will subsequently collapse, hence contributing to drop breakup, as they move to regions of higher pressure. Evidence for the existence of cavitation in the Kodak disperser is the occurrence of pitting on the upstream face of a second orifice plate when installed. Other evidence derives from consideration of the cavitation number

$$\sigma = \frac{P_{exit} - P_v}{\frac{1}{2}\rho \overline{U_o}^2}. \tag{10}$$

Experiments on water flow through an orifice (Yan and Thorpe, 1990) show cavitation occuring when $\sigma < 1.5$ for P_{exit} measured 15 pipe diameters downstream of the orifice and an orifice/pipe diameter ratio equal to the present case ($d_o/D = 0.2$). Values of σ calculated for the Kodak disperser, assuming that P_{exit} is atmospheric (free discharge), are about two orders of magnitude smaller than 1.5; this gives strong additional evidence for the presence of cavitation.

6. Design ideas

6.1 Idea #1 (Remove upstream recirculation zone)

The converging flow into the orifice generates a recirculation zone in the corner just before the orifice plate. The idea was to exclude this "wasted" recirculation from the flow field by altering the boundary shape to follow approximately (as a straight line) the outer converging streamline. Because energy was not being expended driving the recirculation in the proposed new configuration, it was thought that a given applied pressure drop would produce a higher flow rate (hence larger shear rates, more downstream turbulence, and a smaller mean droplet size). However, *Fastflo* numerical calculations showed the opposite effect due to increased wall friction. Idea #1 was therefore abandoned in favour of #2 below.

6.2 Idea #2 (Rounded orifice)

One method of increasing the flow rate for a given pressure drop without the above wall friction penality is to "round" the upstream corner of the orifice. Perry's Chemical Engineers' Handbook shows that a rounded orifice will result in about a 20 per cent increase in flow rate for the same pressure drop.

6.3 Idea #3 (Longer orifice)

In Section 5.2, we estimated that a turbulent boundary layer growing from the orifice entry would fill an orifice of 10–20 diameters in length. Since increasing the amount of turbulence should promote droplet breakup, it seems desirable to use a longer orifice than the minimum 3 mm required to maintain the structural strength of the device. A longer orifice will result in increased wall friction; however the work done against friction is only about 10 per cent (based on an orifice 10 mm long and 1mm diameter) of the work done in accelerating the flow entering the orifice, and should not be a limiting factor.

The use of two closely spaced short orifices, rather than one long orifice, was discussed. It was not clear whether one configuration has an advantage over the other. However, two orifices spaced 1 mm apart, say, may conceivably provide less resistance to flow. In view of the discussion above, the effect is likely to be small, but even small increases in droplet breakup by increased flow rate (at given ΔP) are advantageous.

6.4 Discussion — second orifice plate

Use of a second orifice downstream of the first offers the prospect of additional droplet breakup since it represents an additional pass through a high shear/elongation zone. *Fastflo* calculations predicted that additional high shear rates are indeed present at a second orifice placed 7 mm downstream, and that about 80 per cent of the pressure drop occurs across the first orifice plate. The extra plate will increase the pressure in the region near the front face which will promote the collapse of cavitation bubbles (see Section 5.3). Any additional droplet breakup achieved through exposure to extra strain rate, turbulence, and cavitation must be balanced against the cost of generating the extra pressure drop required.

6.5 Idea #4 (Multi-hole orifice)

Replacing a single hole orifice (diameter d_o) with a number of smaller holes will increase the rate of strain which, in turn, will tend to produce smaller droplets. If the total cross-sectional area is unchanged then the orifice velocity is also unchanged for a fixed pressure drop (ignoring any differences in discharge coefficient resulting from the changed orifice geometry). For four holes, the diameter of each is then $\frac{1}{2}d_o$. In that case G is increased by a factor of two. However turbulence in the orifice, which can also contribute to droplet breakup, will be reduced as Re_o approaches sub-transitional values (constant velocity, reducing d_o).

7. Conclusions

The key issue is how to best use the fixed "budget" of available pressure drop to maximise the influence of factors which promote droplet breakup. These factors include laminar shear/elongation, turbulence, and cavitation. However, the relative importance of these factors is not known in the present application so it is not possible to be more quantitative. Nevertheless, some design ideas have been presented based on simple calculations and qualitative argument. A more detailed study could involve particle tracking methods to trace the history of a representative number of drops within the flow with rules for drop breakup (or otherwise) to be applied using the local conditions encountered by each drop at every time step.

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