LIQUID/LIQUID VISCOUS DISPERSIONS WITH A SMX STATIC MIXER

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Abstract: A SMX static mixer was used to disperse highly viscous Newtonian liquids into less viscous, Newtonian and non-Newtonian fluids without surfactants. The investigation covered the effect of the dispersed phase volume fraction, the viscosity ratio between the phases, the mixer length and the power draw. The flow regime was kept laminar in all the experiments with a dispersed phase content up to 25% and viscosity ratios ranging from 1 to 400. The experimental conditions allowed neglecting the affect of coalescence mainly because of the high viscosity of the continuous phase. The effect of the dispersed phase viscosity and volume fraction on the average drop size was quantified. The size distributions were obtained by image analysis. Using the 'process viscosity' concept, it was possible to collapse all the average diameters on a single master curve by using the energy consumption in the mixer as the shift variable between the experiments. A performance comparison against the Kenics mixer was made using literature results. The results showed that the SMX mixer is much better adapted to the dispersion task mainly due to its internal structure.

Keywords: SMX static mixer; multiphase flow; liquid–liquid dispersion; laminar flow; Newtonian; non-Newtonian.

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INTRODUCTION

Most inline liquid–liquid mixers are either colloidal mills or rotor stators, and the choice between these two technologies is mainly based on the required dispersed phase droplet size to achieve. With viscous or highly viscous fluids, these mixers are almost impossible to use because of their intrinsic design and operating mode that generate either excessive torque or significant viscous dissipation yielding local heating effects. One suitable alternative is to use static mixers, especially the designs well adapted to laminar flows. In the case of liquid–liquid dispersions, the knowledge required to select a particular model or simply determine the specification of a future installation is poorly documented. Most of the available investigations have focused on describing process debottlenecking, ways of getting higher yields in particular applications, and other types of process improvements (e.g., Craik *et al.*, 2003; Benz *et al.*, 2001).

The work of Grace (1982) is considered as the seminal contribution for liquid–liquid dispersions in static mixers in the laminar regime. According to Grace's work, the effect of the flow field type on a single drop over a wide range of viscosity ratio can be expressed by means of the capillary number, namely

$$Ca = \frac{\eta u}{\sigma} = \frac{\tau}{(\sigma/L)} \tag{1}$$

where τ is the viscous stresses on the drop and (σ/L) is the drop resistance to rupture by surface tension (capillary) forces. The critical capillary number is defined as the value that must be reached in a flowing fluid phase in order to break up a dispersed drop of a given viscosity. Following this work, it was further shown that breaking drops of viscosity four times that of the continuous phase and submitted to a simple shear field required longer times but not higher Capillary numbers (Bentley and Leal, 1986). This observation is important since it demonstrates that it is always possible to break a drop provided the deformation or the deformation time is long enough (Stone, 1994).

Time effects are of prime importance in dispersion and even more in static mixers where the geometry generates chaotic flow perturbations (Li *et al.*, 1996). The time required for rupture of a given drop at equilibrium deformation can be estimated with

$$t_{\text{rupture}} = \frac{\eta_c}{(\sigma/D_{\text{drop}})} \tag{2}$$

At very high viscosity ratios, $t_{rupture}$ can reach very high values (Grace, 1982; Bentley and Leal, 1986). Time has then a major influence on the final dispersion and its effects must be considered at the early stages of process design.

The flow type, defined as the ratio of the deformation (D) to the rotational component (Ω) of the flow field (Manas-Zloczower, 1994), is also known as an important parameter governing the breaking mechanism:

$$\alpha = \frac{|D|}{|D| + |\Omega|} \text{ where } D + \Omega = \frac{1}{2}\left[(\nabla v + \nabla v^T) + (\nabla v - \nabla v^T)\right] \quad (3)$$

The value of α is well suited for representing the type of two-dimensional flow used: $0 =$ pure rotational; $0.5 =$ simple shear; $1 =$ pure extensional. Depending on the mixer geometry, any intermediate value between 1.0 and 0.0 is possible. While this parameter cannot be used to compare completely different geometries like an agitated tank and a static mixer (Astarita, 1979), it is still useful to compare similar geometries and various operating conditions in the same mixer. Using numerical simulations, such a comparison has been established successfully for different static mixer models (e.g., Rauline *et al.*, 1998, 2000) and mixing tanks (De la Villéon *et al.*, 1998).

It was experimentally demonstrated that a constant deformation flow (α constant) combined to a decreasing shear rate could lead to the breakup of the dispersed phase. The same size reduction effect was also demonstrated by varying the flow type (α) and the shear rate at the same time, again highlighting the importance of the transient conditions as the ones found in static mixers. From the fundamental contributions of Grace, the conclusions are (Stone and Leal, 1989a, b):

- For a given stretching-sudden relaxation process, the maximum stretching required to rupture the drop at relaxation is widely superior to the maximum stable deformation observed in the critical capillary number experiments. This required stretching at breakup is an increasing function of the viscosity ratio.
- Steps in shear rates during a drop deformation experiment lead to the drop rupture without any important stretching. The breakup is attributed to surface tension effects.
- To allow for capillary instabilities effects to play a role in a filament rupture, the ratio $L_{filament}/R_{filament}$ must be larger than 20.
- Combining and alternating flow types (different α) can lead to drop rupture even though the same flow in stable conditions would not result in such a breakup. This latter observation is of prime importance. It allows for a simple shear flow to break up high viscosity droplets while stable conditions would not.

These conclusions, while having been obtained with single drop experiments, can be directly related to static mixers and their application to dispersion processes. They also partly explain why this type of mixers can effectively reach low average sizes without the typical high shear generated in rotating equipment such as colloidal mills and rotor stators.

The reality of liquid–liquid dispersion is so that millions not to say billions of drops are involved at the same time. The simultaneous rupturing, colliding and coalescence generate a tremendously complex hydrodynamic flow process from an engineering standpoint. Using a large amount of energy in turbulent regime is usually seen as the easiest (and probably the most intuitive) solution provided that the viscosity is sufficiently low (Legrand *et al.*, 2001). The laminar regime is not usually considered as an efficient means for a size reduction process, although Grace (1982) did carry interesting experiments with the Kenics mixer. His ideas were to put in evidence the fact that transient conditions generated in the static mixers could be modeled with abrupt changes in flow conditions. Without ever reaching the critical capillary number, the variations in shear rate effectively led to droplet breakup.

The SMX mixer geometry is very different from that of the Kenics. The SMX shape was used in a dispersion work by Mutsakis *et al.* (1986) who clearly showed that this mixer geometry exhibits both effective shear and elongation to efficiently rupture drops of oil dispersed in a water phase. The major limitation of this research was the restriction of the viscosity ratios to values below 1.

An analogy can be drawn between static mixers and impeller-based mixing in stirred vessels, where equivalence can be derived between the mixer length (or alternatively the number of elements) and the processing time in tanks. The longer the mixer, the better the mixing provided. It has been known for a while that the effect of the number of mixing elements can be modelled with (Middleman, 1974):

$$\frac{D_{32}}{D_{32(N\max)}} = C_1 + C_2 e^{(-N * C_3)} \quad (4)$$

Here, the value of $D_{32(N\max)}$ is simply the minimum size achieved with the maximum number of elements used in the experiments. The values of constants C_1 to C_3 obtained with a Kenics (Middleman, 1974) and an in-liner mixer from Lightnin (Al-Taweel and Walker, 1983) are presented in Table 1.

In parallel with equation (4), the behaviour of the size reduction in the mixer can be explained by a kinetic phenomenon. With the dispersed phase flowing across the static elements, the drops break in a repetitive fashion until equilibrium is reached between the flow forces needed to rupture the drops and the drops resistance due to Laplace pressure. The point where this equilibrium is reached is a function of the dissipated energy and the liquid system involved. The distance to establish the equilibrium can be directly compared to the mixing time in tanks. During the design of a new process application, it is hence of prime importance to evaluate the required number of elements properly.

Table 1. Experimental values of constants C_1, C_2 and C_3 [equation (4) from Middleman, 1974] with conditions.

	Middleman (1974)	Al-Taweel (Al-Taweel and Walker, 1983)
C_1	0.95	1.0
C_2	5.1	4.9
C_3	0.40	0.19
Mixers	3, 6, 21 Kenics	4, 8, 12, 16, 24, 36 in-liner
Fluids	Benzene in water	Kerosene in water
Volume fraction	0.5–1%	1%
Interfacial tension	4 mN m^{-1}	—