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## A COMBINED CRITERION TO IDENTIFY MIXING PERFORMANCE FOR THE BLENDING OF NON-NEWTONIAN FLUIDS USING A KENICS KM STATIC MIXER

### ZŁOŻONE KRYTERIUM DO OKREŚLANIA SKUTECZNOŚCI MIESZANIA PŁYNÓW NIENEWTONOWSKICH ZA POMOCĄ MIESZALNIKA STATYCZNEGO KM KENICS

#### Abstract

Planar Laser Induced Fluorescence (PLIF) has been used to determine the mixing performance of KM static mixers for the blending of Newtonian fluids (aqueous glycerol solutions) and time-independent non-Newtonian fluids (aqueous carbopol 940 solutions), whose rheology shows Herschel-Bulkley behaviour. The effect of number of mixing elements, fluid rheology and apparent viscosity ratio for two-fluid blending have been investigated at constant mixture velocity of 0.3 m s<sup>-1</sup>. For two-fluid blending, the addition of a high viscosity stream into the lower viscosity main flow causes very poor mixing performance, with unmixed spots visible on the PLIF image. Determination of log variance and average striation thickness revealed conflicting trends. A combined area based method allows presentation of a criterion which combines aspects of both scale and intensity of segregation, which can be used in combination with conventional approaches.

*Keywords: scale and intensity of segregation, mixing performance, PLIF, non-Newtonian fluid blending, static mixer*

#### Streszczenie

Fluorescencja wywołana promieniem lasera planarnego (PLIF) jest wykorzystywana do określania poziomu skuteczności mieszania przez mieszalniki statyczne KM łączące płyny newtonowskie (wodne roztwory gliceryny) i niezależne od czasu płyny nienewtonowskie (wodne roztwory carbopolu 940), których reologia wykazuje zachowanie Herschel – Bulkleya. Zbadano wpływ liczby elementów mieszających, reologii płynu i stosunku lepkości pozornej dla mieszaniny dwu-płynowej przy stałej prędkości mieszania wynoszącej 0,3 m s<sup>-1</sup>. W przypadku mieszaniny dwu-płynowej dodanie strugi o wysokiej lepkości do głównego strumienia charakteryzującego się niską lepkością, powoduje niską skuteczność mieszania i występowanie widocznych w obrazie PLIF niejednorodnych plam. Określenie wariancji log i średniej grubości bruzdowania wykazało sprzeczne trendy. Metoda połączona pozwala na zaprezentowanie kryterium łączącego aspekty zarówno skali, jak i intensywności segregacji i może być używana wraz z metodami konwencjonalnymi.

*Słowa kluczowe: skala i intensywność segregacji, skuteczność mieszania, fluorescencja wywołana promieniem lasera planarnego (PLIF), mieszanie płynu nienewtonowskiego, mieszalnik statyczny*

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## 1. Introduction

Laminar mixing, in particular for the blending of non-Newtonian fluids, is ubiquitous in many industries including food, home and personal care, catalyst and plastic manufacture. Applications include the blending of concentrated solid-liquid slurries, polymerizations and the dissolution of solids or surfactants into liquids to form gels or complex surfactant/fluid phases. Within the industrial context, mechanistic understanding of laminar mixing has initially focused on stirred vessels. This is a challenging research topic due to the complexity of the resultant flow fields and combination of mixing mechanisms present, including chaotic mixing. With the ultimate aim of being able to determine mixing quality *a priori*, researchers have performed a combination of experimental and numerical studies on chaotic mixing in stirred tanks [1]. Experimental work has focused on the use of optical flow diagnostic methods such as particle image velocimetry (PIV) or (planar) laser induced fluorescence (PLIF) [2] on transparent systems, whilst modeling has involved direct numerical simulations (DNS) of the Navier-Stokes equations, as well as other forms of Computational Fluid Dynamics (CFD) [3]. Recent work has investigated different aspects of non-Newtonian blending in stirred vessels, focusing on yield stress fluids [4]. Whilst this approach has raised understanding from an empirical to a semi qualitative level, it has highlighted major differences in the mixing behaviour between Newtonian and non-Newtonian fluids.

Whilst stirred vessels remain the workhorse of the chemical industry, the drive towards continuous processing, with consequent sustainability improvements due to reductions in inventory and plant footprint, requires equivalent understanding of non-Newtonian blending within inline static (motionless) mixers. These have been used in industry since the 1950s and design information for the blending of Newtonian fluids is in the public domain e.g. [5]. However, the blending of non-Newtonian fluids is complicated by a non-linear relationship between the applied shear stress and the measured shear rate within the fluid. Newtonian design equations rely on a linear coupling between these quantities, expressing mixing quality relationships in terms of a pipe-averaged shear rate, which is related to the pressure drop per unit length, a measure of the energy input to the fluid to obtain the required mixing [5]. Clearly, this approach is potentially flawed for non-Newtonian systems and must be carefully checked before it is used on real industrial systems. Lagrangian methods have been employed to determine the mixing quality in laminar flow, yet work on non-Newtonian systems has generally focused on pressure drop measurements e.g. [6], with only a few recent studies examining them in more detail [7].

Most work on static mixers has expressed mixing performance in terms of a co-efficient of variance, CoV, (or the log-variance) which is measure of the concentration variance (intensity of segregation) in the final product. Recent work has challenged this approach, considering it necessary to consider the scale of segregation in the mixing process, in terms of the thickness of the striations at the mixer outlet [8]. In certain circumstances this may be a more appropriate measure, or considered in tandem with the CoV.

In this paper, a PLIF based method is described which is used to characterise blending of non-Newtonian fluids in a KM mixer to determine the scale and intensity of segregation as function of number of mixer elements, fluid rheology and apparent viscosity ratio. The method proposed combines scale and intensity of segregation in a single combined criterion which is complementary to existing criteria such as the log variance and striation thickness

[8]. As in previous work [2], the method is based on analysis of images taken from a transverse section across the outlet of mixer, with one fluid phase doped with fluorescent dye.

## 2. Materials and Methods

### 2.1. Experimental Rig and PLIF Setup

Fig. 1 shows a schematic of the experimental rig. A KM static mixer of diameter 12.7 mm (0.5") with either 6 or 12 elements is used. The working fluids are aqueous solutions of glycerol or carbopol 940. The main flow is delivered by a Liquiflo gear pump at  $Q=160 \text{ L hr}^{-1}$ , controlled using a motor drive (Excal Meliamex Ltd). A minor secondary flow, doped with fluorescent dye (Rhodamine 6G) is injected using a Cole-Palmer Micropump (GB-P35) at  $Q=24 \text{ L hr}^{-1}$ . The flow rate is monitored using an electromagnetic flow meter (Krohne). To enable flow measurements to be made using PLIF, which requires optically transparent materials, a T-piece is placed at the end of the pipe which has a glass window inserted perpendicular to the axis of the main pipe. A glass pipe section upstream of the T-piece provides optical access for the laser sheet, provided using a 532 nm Nd-Yag laser (New Wave Solo III). The laser pulses (operating at 7 Hz) are synchronized to the camera (TSI Powerview 4MP 12 bit CCD camera) using a synchronizer (TSI 610035) attached to a personal computer. The camera is equipped with a 545 nm cut-off filter to eliminate reflected laser light, with only the fluorescent light ( $\lambda = 560 \text{ nm}$ ) being captured. The concentration of the minor flow is calculated to obtain an overall concentration in the system  $\sim 0.5 \text{ mg L}^{-1}$  of Rhodamine 6G, within the linear range [9]. Three different fully mixed dye concentrations were used to calibrate the system and confirm linearity, providing a set of linear equations to relate dye concentration to grayscale value on a pixel by pixel basis using MATLAB. The spatial resolution of the measurements was  $10 \mu\text{m pixel}^{-1}$ .

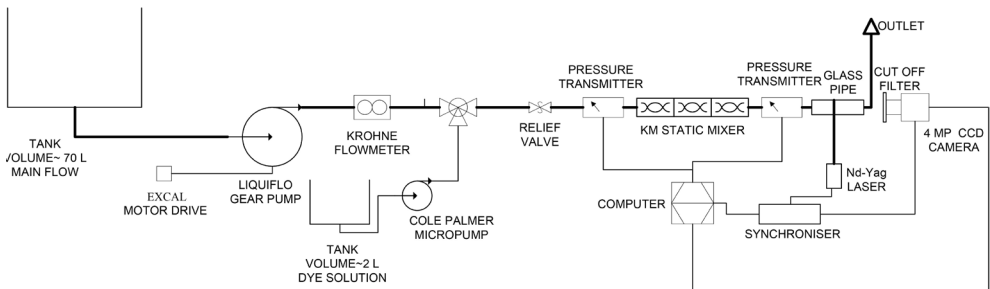


Fig. 1. Schematic of the static mixer rig used for the PLIF experiments

### 2.2. Fluids characterisation

The rheology of the fluids was obtained using a cone and plate rheometer (TA AR1000, TA Instruments) equipped with a 40 mm diameter  $2^\circ$  steel cone. The aqueous carbopol 940 solutions were found to be well represented by the Herschel-Bulkley model (eq. 1) over

a range of shear rates,  $\dot{\gamma}$ , from 0.1 – 1000 s<sup>-1</sup>, where the shear stress,  $\tau$ , is related to  $\dot{\gamma}$ , the yield stress,  $\tau_0$ , consistency index,  $K$  and the power law exponent,  $n$ . The fluid properties are given in Table 1 below.

$$\tau = \tau_0 + K\dot{\gamma}^n \quad \text{for } \tau > \tau_0 \quad (1)$$

Table 1

**Physical properties of the aqueous solutions used in the experiments**

	Density $\rho$ [kg m <sup>-3</sup> ]	Yield stress $\tau_0$ [Pa]	P. law exp. $n$ [-]	Consistency Index $K$ [Pa s <sup>n-1</sup> ]	Viscosity $\eta_A$ (Pa s) (at $\dot{\gamma} = 750$ s <sup>-1</sup> )	pH [-]
Fluid 1: 80% wt Glycerol	1200	-	-	-	0.05	-
Fluid 2: 0.1% wt Carbopol 940	1000	3.7	0.7	0.26	(0.05)	4.5
Fluid 3: 0.2% wt Carbopol 940	1000	25.2	0.42	6.74	(0.25)	5

### 2.3. Experimental conditions

The common range of operational superficial velocities for static mixers is between 0.1-1 m s<sup>-1</sup>. In this work a constant superficial velocity of 0.3 m s<sup>-1</sup> is chosen, determined by practical limitations on the fluid storage volume. Six different experimental conditions have been chosen as shown in Table 2. In addition, for the non-Newtonian experiments, a different fluid (Fluid 3) has been used for the minor (doped) flow than for the main flow (Fluid 2) to examine the mixing of fluids with different rheologies. The apparent viscosity ( $\eta_A$ ) ratio of these fluids is 5 at  $\dot{\gamma} = 750$  s<sup>-1</sup> (Table 1).

Table 2

**Experimental conditions**

No. mixing elements	#1: Newtonian	#2: Non-Newtonian	#3 Non-Newtonian
6 or 12	Main Flow: Fluid 1 Minor Flow: Fluid 1	Main Flow: Fluid 2 Minor Flow: Fluid 2	Main Flow: Fluid 2 Minor Flow: Fluid 3

### 3. Results

#### 3.1. Raw PLIF images

Figure 2 shows a selection of the raw images obtained from the PLIF technique. For the Newtonian blending case (#1), there is a notable reduction in the observed striation thickness when the number of elements is increased (Fig. 2a and 2b), with the overall mixing pattern showing evidence of stretching and folding which is typical for KM static mixers [8]. This is also evident for the non-Newtonian case where both mixed fluids have the same rheology (#2). In both sets of figures, there is a bright region in the bottom right corner of the image when 6 elements are used (Figure 2a, 2c), which suggests some bypassing of the dye stream past the mixer inlet. This phenomenon is exacerbated when the minor flow has a higher apparent viscosity than the main flow in #3. No mixing is observed after 6 elements (Fig. 2e) and the dye remains as a central bright spot. After 12 elements (Fig. 2f), the dye stream has ‘shattered’ into a series of bright spots with some additional evidence of stretching and folding observable.

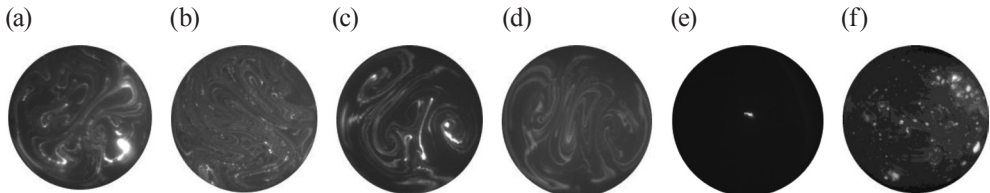


Fig. 2. Raw PLIF images: (a) and (b) show #1 for 6 and 12 elements;  
(c) and (d) show #2 for 6 and 12 elements  
and (e) and (f) show #3 for 6 and 12 elements respectively

#### 3.2. Intensity and scale of segregation

Kukukova et al. [8] proposed an approach which defines segregation as being composed of three separate dimensions. The first dimension is the intensity of segregation quantified by the normalised concentration variance (CoV) or log variance of concentration (LogVa); the second dimension is the scale of segregation or clustering (striation thickness) and the last dimension is the exposure or the potential to reduce segregation. The first dimension focuses on the instantaneous concentration variance, the second on the instantaneous length scales in the mixing field and the third on the driving force for change or the instantaneous rate of reduction in segregation, but this is more important for batch processing. In this work scale and intensity of segregation have been considered. LogVa is determined from the instantaneous concentration variance and is defined in (eq. 2) below, where  $C_i$  is the concentration of the  $i^{\text{th}}$  pixel,  $C_0$  is the concentration in the background and  $C_\infty$  is the concentration assuming perfect mixing across the cross-section:

$$\log \sigma^2 = \frac{1}{N-1} \sum_{i=1}^N \left[ \left( \frac{C_i - C_0}{C_\infty - C_0} \right) - 1 \right]^2 \quad (2)$$

Striation thicknesses were calculated by analysis of the PLIF images in MATLAB. The images were converted into a  $2048 \times 2048$  matrix and an algorithm was written to determine the number of contiguous pixels with the same grayscale value (within a pre-defined tolerance) and thus within the same striation and converted to a length ( $10 \mu\text{m pixel}^{-1}$ ). This was performed on a row by row and column by column basis and the striation distributions were thus recorded and an average value evaluated. Although this method introduces duplication (individual striations identified between adjacent rows or columns are likely to be in the same striation), the trends of the data are valid if the method used is consistent between all images.

Analysis of the images in Fig. 2 using (eq. 2) to obtain LogVa yields the following results shown in Fig. 3a. Unsurprisingly, the results for #1 and #2 are similar, with #3 giving a much worse performance. In contrast, the average striation thicknesses (Fig. 3b) show a different trend, with #3 performing better than #2 for 12 elements, which can be explained by the relatively few striations in #3 skewing the striation distributions, due to lack of mixing. Again, this illustrates the danger of only considering either LogVa or striation thicknesses in determining mixing quality [8].

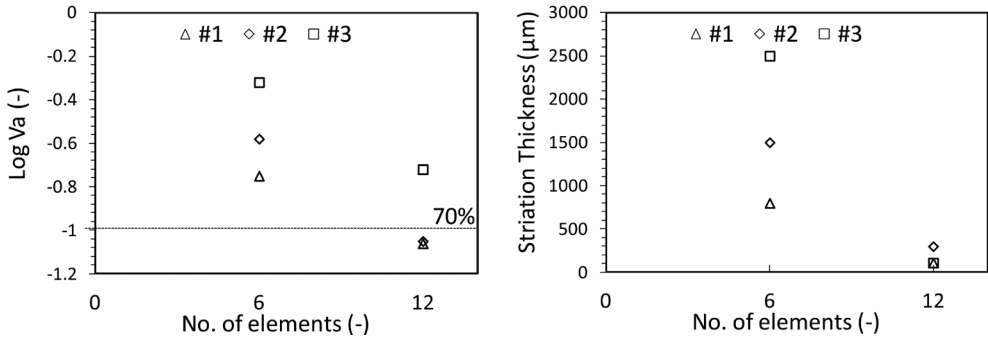


Fig. 3. (a) intensity (log-variance) and (b) scale (striation thicknesses) of segregation for experiments #1, #2 and #3

### 3.3. Area-based analysis combining intensity and scale

In the combined method, the LogVa is used to determine the value of  $C_\infty$  and its corresponding grayscale value  $\bar{G}$ , taken to be ‘perfect’ mixing or 100% mixed. Then, referring to the Fig. 4a, an arbitrary level of mixing, X (%), may be considered, which corresponds to grayscale values of either  $G_{X^-} = (1-X) \times \bar{G}$  or  $G_{X^+} = \bar{G} - (1-X) \times \bar{G}$ , since the squared term in (eq. 2) may have either positive or negative roots. Fig. 4c shows the result of application of this criterion to the raw image in Fig. 4b; areas in the image with a mixing performance greater than an arbitrary percentage are shown in white.

The results of this analysis are shown on Fig. 5 as both a bar graph (Fig. 5a) and cumulative area fraction (Fig. 5b). The Newtonian experiment (#1) gives the best mixing performance, with 39% of the total area containing values of G corresponding to > 90% mixing for 12 elements, reducing to 18% for 6 elements. Experiment # 2 gave a slightly worse performance. However, Experiment #3 gave a very poor result using the combined criterion, re-

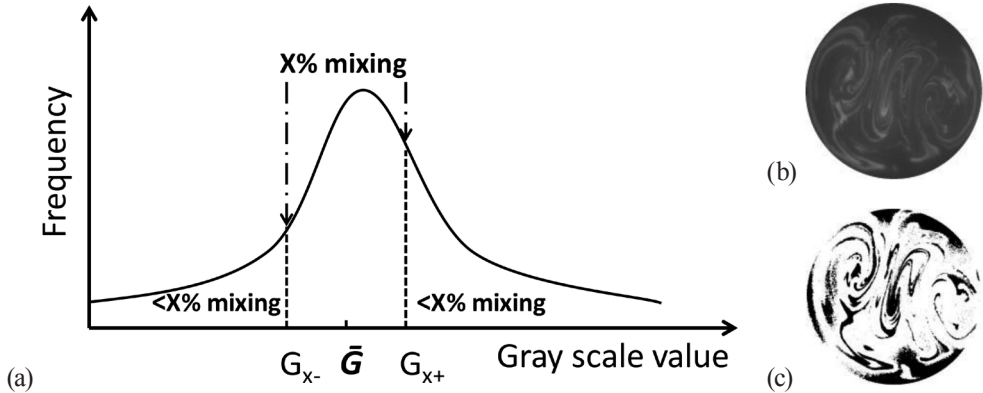


Fig. 4. (a) Identification of regions in the distribution with a given mixing intensity (b) raw image; (c) example of image processing with regions of mixing intensity  $> X\%$  in white

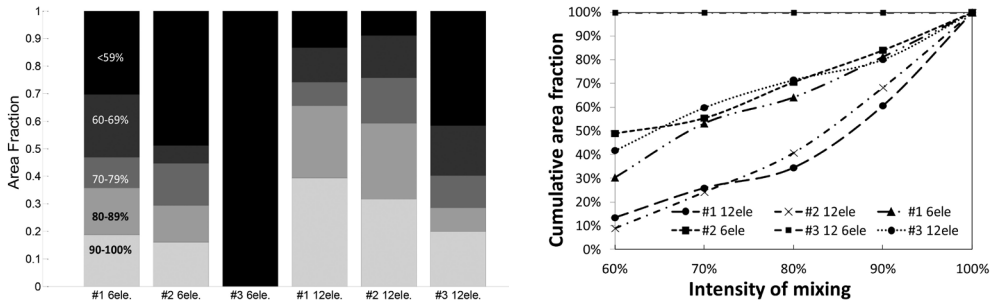


Fig. 5. Results of combined method plotted as (a) bar graph; (b) cumulative area fraction

flecting the images in Figs. 2e and 2f. The entire cross section has values of  $G$  corresponding to less than 50% mixed (as borne out by the log-variance plot in Fig. 3a) when 6 elements are used; the results for 12 elements give close to the same performance as 6 elements in experiments #1 and #2. The cumulative area fraction plot shown in Fig. 5b gives a clearer overall illustration, since improved quality of mixing is observed when the lines shift to the right giving higher total fractions for mixing intensities closer to 100%. The method thus shows promise for characterisation of the mixing performance in terms of area versus intensity.

#### 4. Conclusions

PLIF has been applied to determine the mixing performance of KM static mixers using Newtonian and non-Newtonian aqueous solutions as a function of number of elements and viscosity ratio of the two fluids. Analysis of the data using log variance for intensity of segregation and striation thickness for scale have shown conflicting trends, which is a known problem in the literature [7, 8]. A method is presented which considers the distribution of the cross sectional area with a given intensity of mixing, thus combining attributes of both

intensity and scale enabling them to be considered together. The method shows promise for the evaluation of mixing performance and can be considered as an addition to conventional approaches. Future work will consider the distribution of the size of individual regions with a given mixing intensity, to elucidate the scale of segregation as a function of intensity more precisely.

*FA is funded by an EPSRC DTA studentship and Johnson Matthey.*

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