Striation Thickness Distribution in Split-and-Recombine Mixers in the Stokes Regime

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Abstract

Split-and-recombine (SR) mixers typically operate in the Stokes flow regimes. Numerical and experimental studies have shown that a non-homogeneous distribution of the mixing scales is observed due to the difficulty of ensuring plug flow with viscous liquids. The parabolic velocity profile generated in the duct promotes the formation of larger mixing scales next to the walls and smaller scales in the centre of the duct. The mixing efficiency controlled by diffusion at the molecular scale depends on the non-ideal striation thickness decay. In this work, a simple model to estimate the distribution of mixing length scales in a SR mixer is proposed and compared to Computational Fluid Dynamics simulations. It is shown that the striation thickness of the largest scales in the flow decays at half the rate of an idealised baker's transformation, and that an SR mixer must be 1.81 times longer than an ideal one to achieve complete mixedness. The intensity of segregation is calculated for the largest mixing scales and set as an industrial design criterion for SR mixers. The main novelty of this work is the introduction of a new model to describe the non-homogeneous distribution of mixing scales in SR mixers.
Keywords
Static Mixers, Striation Thickness, Mixing, Intensity of Segregation, Stokes Flow Regime, Multi-level laminating micromixers

Introduction
Static mixers are widely used in laboratory and industrial scale applications to mix two or more fluids in a continuous fashion without the use of moving mechanical elements. These mixers are especially suited for applications where flow rates are too small (e.g. microfluidic applications) or the viscosity is too large (e.g. polymers, detergents, paints) to achieve turbulence-induced mixing in the flow.

The use of these mixers in industry only became widespread after the 1970s, but there are patents prior to this date (Sutherland, 1874; Bakker, 1949; Stearns, 1949; Lynn, 1951; Tollar, 1960; Veazey, 1965). The first patent was issued in 1874, and it describes a multilayer reactor without moving parts used to mix air and gaseous fuel (Sutherland, 1874).

Static mixers can have specific geometries that promote secondary and transverse flow, enhancing the mass and heat transfer. One type of static mixers is the insert-type of multiple stationary parts, called elements. These elements may be blades or conjugated plates that induce changes in the flow. These insets can be helical elements or oblique blades (Ghanem et al., 2014) to ensure the stretching and engulfment of the incoming fluids promoting distributive mixing. Another type of static mixers is based on channels without baffles or other obstacles, such as opposed jets mixers, which are based on the injection of liquid streams as two opposed jets. These mixers where mixing is promoted without external energy and moving parts are commonly called passive mixers. Confined Impinging Jets mixers with a cylindrical geometry (Keuerleber and Pahl, 1970; Malguarnera and Suh, 1977; Ritcher and Macosko, 1978; Lee et al., 1980; Tucker and Suh, 1980; Sandell et al., 1983; Johnson et al., 1996; Johnson and Wood, 2000; Fonte et al., 2015) and T-Jets with a prismatic geometric
(Kockmann et al., 2003; Engler et al., 2004; Hoffmann et al., 2006; Sultan et al., 2012; Mariotti et al., 2018; Mariotti et al., 2019) are two of the most common opposed jets mixers used in the industry. A novel static mixer that extends this concept into a network, the NETmix, was more recently proposed (Laranjeira et al., 2009; Laranjeira et al., 2011). NETmix is based on a network of mixing elements that consist of opposed jets mixing chambers connected by channels. These static mixers are efficient at dynamic flow regimes, where the inertia of jets is key to promote vortices that engulf fluids, which generally occur at operation Reynolds numbers larger than 100.

Mixing in passive mixers at low Reynolds number is completely characterised from the temporal and spatial variations of residence time distribution (Castelain et al., 1997; Aubin et al., 2005; Ghanem et al., 2014) and the striation thickness distribution (Sokolov and Blumen, 1991; Aubin et al., 2005; Ghanem et al., 2014). The design of reactors geometry and the number of static mixing elements are the parameters that most influence the mixing efficiency.

Passive mixers have been used as an alternative to conventional reactors. The potential advantages of static mixers compared to mechanically agitated vessels are the small space requirements, low equipment cost, better energy efficiency for mixing, no moving parts, narrow span of residence times distribution approaching plug flow, good mixing at low shear rates, flexible production of different products in the same device and self-cleaning (Thakur et al., 2003).

The design of static mixers can be tailored for the respective applications. Static mixers have been used for different systems, such as liquid-liquid, gas-liquid and solid-liquid systems with a particular application in chemical, petrochemical, food, pharmaceutical, pulp and paper industries (Thakur et al., 2003). Static mixers are commonly divided into four groups according to their application (Thakur et al., 2003):

- Mixing of miscible fluids working at laminar or turbulent flow regimes;
- Mixing of immiscible fluids with the generation of interfacial area in liquid/liquid, gas/liquid and solid/liquid systems;
- Heat exchange and thermal homogenisation;
- Solids’ blending.

Static mixers first applications focused on mixing of fluids at laminar flow regime conditions. Applications as heat transfer, turbulence and multiphase systems were introduced later (Thakur et al., 2003). The objective of these first applications was to achieve good mixing in the cross-section of a circular tube at laminar flow regime. Elements were installed in a static mixer to promote the redistribution of the incoming liquid stream and then the combination into different layers in a sequence. Mixing in these devices does not rely on the onset of inertial mechanisms, such as turbulent eddies or chaotic motion, but by the reduction of the mixing scales, i.e. elongated lamellar structures created at non-inertial regimes. The generation of thin lamellae enhances the concentration gradients and the molecular diffusion of phases.

Mixing in static mixers can also occur in turbulent flow regime. In these devices, inserts promote turbulence and generate intense radial mixing. Three examples of applications are the mixing of gas at turbulent flow regimes, the blending of polymer melts and aqueous solutions (Ghanem et al., 2014). Mixing performance is not easy to measure at turbulent flow regime. Lemenand et al. (2017) assessed micromixing from chemical probe measurements applied to two phenomenological models: IEM (Interaction Exchange with the Mean) and EDD (Engulfment, Deformation and Diffusion).

In this work, a particular focus is given to single-phase systems in static mixers operating under the Stokes regime.

**Fundamentals of Mixing in Static Mixers at Stokes Flow Regime**

Mixing in the laminar flow regime usually occurs for high-viscosity fluids in the food, coatings, cosmetic, polymer, adhesive and detergent industries. The split-and-recombine (SR) reactor is a passive static mixer widely used in these industries that typically operates at low Reynolds
number. These reactors are based on the split and recombine principle, which consists in splitting the incoming fluid into layers and then recombine them in a different sequence. No external agitation is required to ensure the mixing of liquid streams (Gray et al., 1999; Chen and Meiners, 2004; Hirata and Ohkawa, 2016; Al-Hassan et al., 2021), and the specific geometrical construction of the insert-type configuration promotes the redistribution of fluids. SR mixers are also known as multi-level laminating micromixers. In the last decades, different geometries that promote the splitting and recombining of passive SR mixers have been tested (Parsa and Hormozi, 2014; Hossain and Kim, 2015; Habchi et al., 2018; Habchi et al., 2019).

Mixing in SR mixers working at laminar flow regime can be described by one of the most well-known one-dimensional dynamical systems transformation, the baker’s transformation (Ottino and Wiggins, 2004). Figure 1 shows a schematic representation of baker’s transformation. Square A, which consists of two strips (white and black), is split in two rectangles with a length $L$ and a height $L/2$. Then these rectangles are rotated and stretched, resulting in two rectangles with a length $L/2$ and a height $L$. The fusion of elements forms a new part with 4 strips, which results from the combination of the mechanism of splitting and stretching.

![Figure 1 The Baker’s transformation.](image)

The main contribution of the baker’s transformation to the mixing efficiency is the increase of the interfacial area between fluids (Ghanem et al., 2014; Hossain and Kim, 2015). The lamellar structure generated from the cutting and re-stacking mechanisms promotes the reduction of diffusion length (Aoki et al., 2011), resulting in enhanced diffusion through the fluids interface.
The repetition of the baker’s transformation promotes the creation of a multi-lamination square. Figure 2 shows a contour map of a successive implementation of baker’s transformation in a system of two strips (Figure 2a). This figure shows that the repetition of iterations determines the efficiency of the mixing degree. The result is the formation of 4, 8 and 16 strips, as shown from Figure 2b to Figure 2d. Considering that the number of transformations is described by \( n \), the number of strips increases \( 2^n \) (Wiggins and Ottino, 2004) and thus, the thickness of each strip is given by \( \delta = L/2^n \) where \( L \) is the unit length.

Ruijin et al. (2017) compared the mixing performance of different geometrical configurations. Results show that passive mixers based on baker’s transformation have better mixing efficiency at low Reynolds numbers since, at these conditions, the formation of multi-lamellae promotes diffusion mixing. At higher Reynolds number, mixing is dominated by convective mechanisms.

The evolution of the interfacial area and the striation thickness decay are commonly used as the metrics to assess the quality of mixing. The interfacial area is promoted by the stretching mechanism and the thinning of striation thickness that accelerates the interdiffusion at molecular scale. Stretching is related in a fundamental way to the Lyapunov exponent, \( \Lambda \), which is given by

\[
\Lambda = \lim_{n \to \infty} \frac{1}{n} \ln (\lambda(X, t))
\]  

(1)

where \( \lambda \) is the stretching of a particle in the initial position \( X \) and time \( t \). The stretching factor for the baker’s transformation is related to the increase in the number of striations, \( 2^n \), and so \( \lambda \approx 2^n \), which is the inverse of the thinning at each transformation. Therefore, according to Equation (1), SR mixer has a characteristic Lyapunov exponent equal to \( \ln(2) \).
The reproduction of the baker’s transformation in mixers only occurs for a specific 3D stationary continuous mixer configuration, which is thought to approach the separation/stacking mechanism. Carrière (2007) proposed a geometry that is a network of converging "T"s where two fluids are injected in a mixing chamber. Figure 3 shows a schematic representation of this mixer. Each "T" plays an important role in the stretching and re-stacking of the two streams, and thus it corresponds to an element of the static mixer (Ottino, 1989). On the other hand, cutting and splitting take place at the outlet of each mixing element (Carrière, 2007). Carrière (2007) studied the mixing dynamics from CFD simulations in the Stokes regime, intending to mimic the baker’s maps. Carrière (2007) also compared the theoretical Lyapunov exponent with CFD results, demonstrating that the result is $\Lambda = 0.68$, which is close to the value predicted by the baker’s transformation, $\ln(2)$.

Figure 3 SR mixer proposed by Carrière (2007).
Schoenfeld et al. (2004) also validated the SR mixing concept from numerical and experimental results. The mixing performance was investigated for a Reynolds number range from 1 to 100. CFD simulations show that for $Re < 15$, the multi-lamination process has a similar evolution to the idealised theory in Figure 2. The same research team proposed a theoretical model based on 1D diffusion equation, which predicts the hydrodynamics of the flow, i.e. the flow splitting and recombining (Hardt et al., 2006). In this model, the lamellae are equally spread over the width of a mixing channel, according to baker’s transformation. The main advantage of this model is that it is numerical diffusion free, avoiding the overprediction of mixing speed.

Mixing in SR mixers was quantified by Neerincx et al. (2011) from experiments using two coloured epoxies, which were polymerised after flow and the mixer was sliced to visualise the flow structures. A specific SR mixer configuration, the Dentlncx mixer, was used and consisted of a splitting serpentine channel. Results show that the formation of strias in these reactors exactly follows the baker’s transformation. Nevertheless, the mapping of the concentration distribution shows a non-uniform striation thickness at the outlet of each element.

Schoenfeld et al. (2004) and Nimafar et al. (2012) compared a “T” element and a duct with square cross-section. In these geometries, the ideal striation thickness decay, $\delta = 1/2^n$, is not attained because a plug flow of viscous fluids is not ensured in the mixing elements. The actual flow velocity profile is set by the no-slip condition at the walls and the viscous stresses.

One of the main problems of mixing viscous fluids in passive SR mixers is the pressure drop because it requires high energy consumption. Habchi et al. (2019) studied the mixing quality and the pressure drop in passive SR mixers with the same geometries described in Gray et al. (1999) and Chen and Meiners (2004). The performance of these mixers was compared with a plain duct. Results showed that SR mixers enhance the concentration gradients promoting molecular diffusion, guaranteeing better performance. These geometries are associated to lower pressure drops compared to a plain duct because fluids flow with half of the inlet velocity over much of the reactor length due to the splitting of the channel.
In this work, a new model for the striation thickness evolution is proposed considering that two Newtonian fluids will be divided into strips of equal flow rate for each fluid but maintaining the velocity profile in the channel. This generic model is able to describe the mixing of fluids at unsymmetrical flow conditions, i.e. when the two jets have different flow rates. The validation of this model is made by comparison with CFD results.

The striation thickness evolution has an impact on the mixing degree because the diffusion at molecular scales is controlled by the rate of evolution of the mixing scales and the respective rate of interfacial area generation. Thus, the mixing efficiency is strongly dependent on the reduction of scales in SR mixers. The validation and consequent implementation of the striation thickness decay model are quite useful for the design of SR mixers as industrial mixing units.

The main novelty of this work is the introduction of a new model to describe the striation thickness decay in SR mixers. The mixing structures in these mixers have been previously validated from experimental and numerical results in other scientific works (Schönfeld et al., 2004; Nimafar et al., 2012). However, they are described by a theoretical framework for the first time. An explicit equation describing mixing is proposed as a process intensification methodology that can be used in a wide range of industries as food, cosmetic, coatings, polymer, adhesive and detergent.

**Analytical Model Development**

The geometry addressed in this work is similar to the one proposed by Carrière (2007), which has been designed to reproduce as closely as possible the mixing properties of a baker’s transformation. This geometry consists of a network of converging and diverging “T”s. The number of transformations in this geometry corresponds to the number of mixing elements, i.e. to the number of converging “T”s. It was considered that the first mixing element coincides to the one that transforms two in four strips. Therefore, the number of strias generated in each mixing element is described by $2^{n+1}$.
As mentioned before, this striation thickness decay law in a real mixer is not feasible due to the impossibility of ensuring plug flow of viscous fluids in the mixer channels. In practice, SR mixers with rectangular cross-section will develop a velocity profile that can be calculated in its dimensionless form

\[
v(\xi^*, \psi^*) = \frac{\sum_{i=0}^{\infty} \sum_{j=0}^{\infty} \frac{\sin(i\pi\xi^*) \sin(i\pi\psi^*)}{4i^2j^2}}{4 \sum_{i=0}^{\infty} \sum_{j=0}^{\infty} \frac{i^2j^2}{\pi^2i^2j^2}(\beta_i^2i^2 + j^2)}
\]  

(2)

where \(0 \leq \xi^* \leq 1\) and \(0 \leq \psi^* \leq \beta_c\) are the dimensionless spatial coordinates and \(\beta_c\) is the aspect ratio of the channel given by the ratio between height and length of the cross-section channel (Spiga and Morino, 1994); \(\xi^*\) and \(\psi^*\) are defined from the ratio between spatial coordinates \((x, y)\) and the edge length in the respective direction, i.e. \(\xi^* = x/l\) and \(\psi^* = y/h\). All spatial coordinates have been made dimensionless by a characteristic length \(L\) of the channel, and the velocity is divided by its mean value in the cross-section in the channel. This velocity profile will generate, after each mixing element, a non-homogeneous distribution of striation thicknesses.

The striation evolution after each mixing element can be estimated for steady state flows by performing the following mass balance

\[
\int_{A_i} \rho v(\xi^*, \psi^*) dA = \rho q_i
\]

(3)

that considers that the two fluids will be divided into \(i\) striations of equal flow rate for each fluid, \(q_i\), but accounting for the actual velocity profile in the channel (Equation (2)). It was assumed in this work that both fluids have the same physical properties. This analytical model also considers that both fluids are Newtonian, and thus its implementation is only valid for processes that involve fluids whose viscosity is independent of the shear rate.

The position of the striations interface, \(e_i\), after a transformation \(n\) at a given position \(\psi^*\), in the normal direction of the orientation of strias, can be estimated from
\[
\int_{\varepsilon_i}^{\varepsilon_i+1} v(\xi^*, \psi^*) d\xi^* \Delta \psi^* = \frac{r_s a}{2^{n+1}(r_s + 1)} \int_0^1 v(\xi^*, \psi^*) d\xi^* \Delta \psi^* \tag{4}
\]

where \( i = 1, 2, ..., 2^{n+1} \) according to the baker transformation, where the positions of interfaces for \( i = 0 \) and \( i = 2^{n+1} \) are \( \varepsilon_0 = 0 \) and \( \varepsilon_{2^{n+1}} = 1 \). \( \Delta \psi^* \) represents an infinitesimally small spatial coordinate in the \( \psi^* \) direction and \( r_s \) is the flow rate ratio between the two phases, 1 and 2, given by

\[
r_s = \frac{q_1}{q_2} \tag{5}
\]

The parameter \( a \) in Equation (4) is

\[
a = \begin{cases} 
0 & \text{if } i \text{ is even} \\
1 & \text{if } i \text{ is odd} 
\end{cases} \tag{6}
\]

The solution of Equation (4) gives the thickness of each stria, which can be calculated by

\[
\delta_i = \varepsilon_i - \varepsilon_{i-1} \text{ for } i = 1, 2, ..., 2^{n+1}. 
\]

A 1D simplification of Equation (2) assuming a parabolic profile can be considered to predict the velocity profile in \( \xi^* \) direction. A system of two infinite plates was assumed by Bird et al. (1924) to approximately describe the 1D velocity profile in a square duct, which is given by

\[
v(\xi^*) = 6\xi^* (1 - \xi^*) \tag{7}
\]

Equation (7) can be used to obtain a simpler and explicit way of calculating the distribution of striation thicknesses. Figure 4 shows the comparison between the velocity profile in a square chamber at \( \psi^* = 1/2 \) and the approximation to a fully parabolic profile. Equation (7) can be used as a simplification of Equation (2) since the profiles are similar. Both profiles in Figure 4 have been divided by \( \int_0^1 v(\xi^*) d\xi^* \) for comparison. This simplification was verified in this work by the use of a spatially resolved computational model of the flow.

The combination of Equations (4) and (7) results in

\[
2\varepsilon_{i-1}^3 - 3\varepsilon_{i-1}^2 + 2\varepsilon_i^3 + 3\varepsilon_i^2 = \frac{r_s a}{2^{n+1}(r_s + 1)} \tag{8}
\]
that can be further solved to obtain an explicit general solution for the position $\varepsilon_i$ of the interface between the two fluids for a given flow rate ratio $r_5$ and after the mixing element $n$

$$\varepsilon_i = \cos \left( \frac{1}{3} \cos^{-1} \left( 1 + 4\varepsilon_{i-1}^3 - 6\varepsilon_{i-1}^2 - \frac{2^{1-n}r_5}{r_5 + 1} \right) - \frac{2\pi}{3} \right) + \frac{1}{2}$$  \hspace{1cm} (9)

for $i = 1$ to $2^{n+1}$ and $\varepsilon_0 = 0$.

![Image](image_url)

**Figure 4** Velocity profile in the middle of the square channel and the parabolic approximation.

Figure 5 shows the generation of a non-uniform distribution of the striation thickness calculated from Equation (9) and the respective velocity profile. The thinnest lamellae are at the centre of the mixing chamber while the thickest lamellae are next to the walls. This observation is in agreement with the shape of the velocity profile in the duct. According to the velocity profile of a square duct and also shown in Figure 5, the largest lamellae for each fluid 1 and 2 will be the ones closer to the walls of the mixer.

Equation (9) can be further simplified to calculate the thickness decay of the largest strips in the process. Considering that fluid 1 is injected on the left side of the chamber, the decay of the largest lamellae, $\delta_{1,\text{max}} = \varepsilon_1 - \varepsilon_0$, that is, the first lamella formed next to the walls, follows

$$\delta_{1,\text{max}}(n, r_5) = \cos \left( \frac{1}{3} \cos^{-1} \left( 1 - \frac{2^{1-n}r_5}{r_5 + 1} \right) - \frac{2\pi}{3} \right) + \frac{1}{2}$$  \hspace{1cm} (10)
and for fluid 2, the thickness of the largest strip, which is formed next to the opposite wall, is calculated from

$$\delta_{2,\text{max}}(n, r_s) = \cos \left( \frac{1}{3} \cos^{-1} \left( 1 - \frac{2^{1-n}}{r_s} \right) - \frac{2\pi}{3} \right) + \frac{1}{2}$$

(11)

In the particular case of $r_s = 1$, i.e., equal flow rates of both fluids, the largest and smallest strips in the mixer, $\delta_{\text{max}}$ and $\delta_{\text{min}}$, respectively, are given by

$$\delta_{\text{max}}(n) = \cos \left( \frac{1}{3} \cos^{-1} \left( 1 - 2^{-n} \right) - \frac{2\pi}{3} \right) + \frac{1}{2}$$

(12)

and

$$\delta_{\text{min}}(n) = \cos \left( \frac{1}{3} \cos^{-1} \left( -2^{-n} \right) - \frac{2\pi}{3} \right) + \frac{1}{2}$$

(13)

![Image](image_url)

Figure 5 Generation of unequal thickness strips due to the presence of parabolic-like velocity profiles in the channel of the SR mixer (a) $n = 0$; (b) $n = 1$ and (c) $n = 2$.

**Numerical Simulations of the Flow and Advective Mixing**

The physical domain simulated in this work is shown in Figure 6a and consists of an SR mixer with two inlets, one outlet and three mixing elements. The geometry dimensions are summarised in Table 1.
Table 1. Dimensions of geometry dimensions

<table>
<thead>
<tr>
<th>Region</th>
<th>Dimensions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cross-section of square duct</td>
<td>2 x 2 mm</td>
</tr>
<tr>
<td>Inlet pipe length of the 1st mixing element</td>
<td>10 mm</td>
</tr>
<tr>
<td>Mixing element length</td>
<td>4 mm</td>
</tr>
<tr>
<td>Inlet pipe length except for the 1st mixing element</td>
<td>14 mm</td>
</tr>
<tr>
<td>Total length</td>
<td>32 mm</td>
</tr>
</tbody>
</table>

The selection of this geometry was based on the criterion in Carrière (2007), who claims that this configuration promotes the splitting and recombining of fluids in a simple way. The main advantages of this mixer are easy fabrication and implementation, large stretching rates and efficient heat exchange due to the large surface-to-volume ratio. The main disadvantage is the complex fabrication process.

The Finite Volume code ANSYS Fluent v18 was used to solve the dimensionless governing equations that describe the steady laminar flow in the SR mixer,

\[ \nabla \cdot \mathbf{u} = 0 \]  \hspace{1cm} (14)

and

\[ \frac{1}{Re} \nabla^2 \mathbf{u} - \nabla p = 0 \]  \hspace{1cm} (15)

where \( p \) is the dimensionless relative pressure and \( \mathbf{u} \) is the dimensionless velocity vector field.

Fluent has an implementation of steady-state Navier-Stokes equations that in practice simplifies to Equation (15), because the simulations were restricted to Stokes regime (i.e. creeping flow conditions), corresponding to a Reynolds number range \( Re < < 1 \). The Volume-of-Fluid (VOF) method was used to track the interface between two different fluids, 1 and 2, entering from each inlet as

\[ \mathbf{u} \cdot \nabla \phi_1 = 0 \]  \hspace{1cm} (16)
where \( \phi_1 \in [0,1] \) is the local volume fraction of fluid 1. The setting of VOF model in ANSYS Fluent enables the simulation of the purely advective flow of two fluids, neglecting surface tension and diffusive transport. The assumption of no diffusion and no surface tension between liquid phases enables the visualisation of laminar mixing mechanisms, which is the main target that we want to validate in this work. Both fluids are Newtonian and have the same physical properties, i.e. the same density and viscosity.

![Figure 6](image_url)

**Figure 6** (a) Geometrical domain for the CFD simulations; (b) Mesh Distribution for adaptative mesh at the beginning and for the converged solution at \( r_s = 1 \).

Uniform discretisation, capable to completely describe the lamellar structure, would result in an expensive computational grid due to the high number of elements. According to the expression that describes the generation of lamellae, \( i = 1, 2, \ldots, 2^{n+1} \), 16 lamellae are formed at \( n = 3 \). The visualisation of 16 lamellae at the outlet requires the generation of a mesh with an edge \( 2/16 = 0.125 \) mm. This simulation would require approximately \( 6 \times 10^5 \) elements.

In order to overcome this issue, a dynamic grid-adapting algorithm was used to refine the numerical mesh at each iteration only at the interface between the two fluids. The domain was initially discretised with a structured mesh consisting of finite cubic elements with volume
\((L/20)^3\) able to solve the flow field, where \(L\) is the length of edge of the reactor \((L = 2\text{ mm})\). This mesh has initially 616 000 elements. The domain was then adapted at the interface of both fluids resulting in 8 030 827 elements by the end of the simulation. Thus, two maximum levels of refinement were used, which corresponds to finite volume elements with a minimum volume \((L/80)^3\). Figure 6b shows the initial and adapted mesh for the converged simulation at \(r_s = 1\).

Another mesh with no adaptive mesh function was considered in order to test the grid independence. This second mesh has 4 928 000 elements, each one with a volume of \(1.25 \times 10^{-13}\text{ m}^3\). Results show that the velocity profiles at the inlet of each mixing element are well-defined by the initial number of mixing elements of the adaptative mesh.

The imposed boundary conditions at the inlets, outlet and the walls were as follows,

\[
\begin{align*}
\text{Inlet A: } & \quad \mathbf{u} \cdot \mathbf{n} = \frac{r_s}{r_{s+1}}, \nabla p \cdot \mathbf{n} = 0, \phi_1 = 0 \\
\text{Inlet B: } & \quad \mathbf{u} \cdot \mathbf{n} = \frac{1}{r_{s+1}}, \nabla p \cdot \mathbf{n} = 0, \phi_1 = 1 \\
\text{Walls: } & \quad \mathbf{u} = 0, \nabla p \cdot \mathbf{n} = 0, \nabla \phi_1 \cdot \mathbf{n} = 0 \\
\text{Outlet: } & \quad \nabla \mathbf{u} \cdot \mathbf{n} = 0, p = 0, \nabla \phi_1 \cdot \mathbf{n} = 0
\end{align*}
\]  

(17)

The simulations of the flow and purely advective mixing were performed for \(r_s = \{1,2,10\}\). The coupled scheme was set to solve the pressure-velocity equation because it is a robust and efficient single-phase implementation for steady-state flows. Least Squares Cell Based was selected for gradient discretisation, because its accuracy is much superior to cell-based gradient. PRESTO! discretisation for spatial gradients of pressure is accurate for this case because it avoids pressure gradient assumption on boundaries. Third-Order MUSCL accurately discretises the spatial gradients using a combination of the central differencing scheme and second-order upwind scheme. For volume fraction discretisation, Compressive method was chosen, that is a high-resolution difference scheme. The simulations were considered converged for residuals smaller than \(10^{-8}\) for all variables.
This study is only based on numerical data. The validation of CFD results was made from analytical expressions, which are based on the velocity profile previously compared to experimental results in literature (Spiga and Morino, 1994).

Results and Discussion

CFD simulations enable to obtain a realistic distribution of the two liquids inside the SR mixer geometry. Figure 7 shows the contour maps of the volume fraction distribution in cross-sections of the mixer placed before the bifurcation that coincides to the outlet of each converging T. Two phases were injected from the opposed jets at three different flow rate conditions, $r_s = \{1, 2, 10\}$. The dark liquid is injected from the left injector, inlet 1, and the white phase from the right-hand side injector, jet 2. Position $n = 0$ corresponds to the chamber where the jets are impinged. $n > 1$ corresponds to the cross-section right after the converging T of the $i$-th mixing element and before the diverging T of the $(i+1)$-th mixing element.

At $n = 0$, a structure of two lamellae, one white and one black, are observed independently of the flow rate ratio, $r_s = \{1, 2, 10\}$ (Figure 7a to Figure 7c). At this mixing element, the thickness of each lamella is the same for $r_s = 1$. However, the dark phase is observed to become thicker for $r_s > 1$, because the flow rate $q_1$ is larger than $q_2$. Then, this structure leaves the first converging T, where it is split, rotated and injected in another converging T. Four lamella are then generated by injection of these streams, as shown in Figure 7d to Figure 7f. The same procedure is repeated for $n = 2$ and $n = 3$.

Figure 7 shows that the SR mixer performs a baker’s transformation to the flow in the sense that after each mixing element, the number of striations of each fluid follows the sequence $2^{n+1}$. However, as expected, due to the velocity profile in channels, the developed striations do not have the same thickness. For all simulated cases, striations close to the walls remain thicker than the striations at the centre of the chamber. The lamellar structure formation is clearly observed from the contour maps in Figure 7, except after the third element for $r_s = 10$ (Figure 7). At this condition, the flow rate $q_1$ is much higher than $q_2$, and thus thinner.
lamellae of white fluid would be formed at the third element of SR mixers. These structures are not clearly observed from the contour maps because the lamellar thickness is smaller than the size of the smallest possible elements in the grid after adaptation. The insufficient grid refinement results in the break of lamellae, as observed in the contour map of Figure 7. This observation shows the difficulty to simulate the mixing of a passive tracer in an SR system even in the Stokes regimes. The model validation avoids further CFD simulations for a sufficient grid refinement because the analytical expression gives an accurate description of the interface between the two phases. Therefore, the insufficient grid refinement to simulate conditions for \( r_s = 10 \) and \( n = 3 \), evidences that the development of a model to describe the striation thickness decay in SR mixer is of utmost importance to simplify the simulation at these conditions.

CFD results also show that a significant pressure drop inside the reactor is promoted by the flow of high viscous fluids. For \( r_s = 1 \), \( \Delta p = 4.87 \times 10^5 \) Pa, \( \Delta p = 7.727 \times 10^5 \) Pa for \( r_s = 2 \) and \( \Delta p = 3.06 \times 10^5 \) Pa for \( r_s = 10 \). These values were also validated from the Poiseuille equation for rectangular channels.

Figure 8 compares CFD results and the model that describes the position of each lamella interface at each mixing element (Equation (9)). These figures show a good agreement between the model and CFD results, thus obtaining a linear relationship between both results. The validation for \( r_s = 1 \) and \( r_s = 2 \) was made for \( n = 2 \) and \( n = 3 \). For \( r_s = 10 \), the comparison between CFD results and the model is only considered up to the second mixing element because for \( n = 3 \), the insufficient grid refinement does not enable an accurate determination of the striation thickness. The good agreement between CFD results and the analytical model also proves that the 1D simplification of velocity profile in a square duct is a good approximation of Equation (2).
Figure 7 Striation Thickness Distribution from CFD simulations for (a) $r_\gamma = 1, n = 0$; (b) $r_\gamma = 2, n = 0$; (c) $r_\gamma = 10, n = 0$; (d) $r_\gamma = 1, n = 1$; (e) $r_\gamma = 2, n = 1$; (f) $r_\gamma = 10, n = 1$; (g) $r_\gamma = 1, n = 2$; (h) $r_\gamma = 2, n = 2$; (i) $r_\gamma = 10, n = 2$; (j) $r_\gamma = 1, n = 3$; (k) $r_\gamma = 2, n = 3$; (l) $r_\gamma = 10, n = 3$. 

\[
\begin{align*}
\gamma & = 1 \\
n & = 0 \\
r_\gamma & = 2 \\
n & = 10 \\
r_\gamma & = 1 \\
n & = 1 \\
r_\gamma & = 2 \\
n & = 2 \\
r_\gamma & = 10 \\
n & = 2 \\
r_\gamma & = 1 \\
n & = 3 \\
r_\gamma & = 2 \\
n & = 3 \\
r_\gamma & = 10 \\
n & = 3
\end{align*}
\]
Figure 8 Parity plots comparing of the interface positions obtained from the analytical model and from the CFD simulations for different values of $r_s$ after (a) 2 mixing elements and (b) 3 mixing elements.

The validation of the position interface by Equation (9) enables to predict the maximum and minimum striation thickness decay from Equations (12) and (13). The rate of the thickness decays of the largest and smallest lamellae for $r_s = 1$ are plotted in Figure 9, showing that the thickness decay of the maximum mixing scale is described by approximately $2^{-n/2}$ and for the minimum mixing scale by $2^{-n}$.

The comparison between the idealised model and the expressions for the maximum and minimum lamellae can be made from Lyapunov exponent. From the mathematical definition, the Lyapunov exponents calculated for the largest and smallest lamellae are, respectively,

$$\Lambda_{\text{max}} = \lim_{n \to \infty} \frac{1}{n} \ln (\delta_{\text{max}}^{-1}(n)) \sim \frac{\ln(2)}{2}$$

$$\Lambda_{\text{min}} = \lim_{n \to \infty} \frac{1}{n} \ln (\delta_{\text{min}}^{-1}(n)) \sim \ln(2)$$

(18)

where $\delta_{\text{max}}(t) \sim 2^{-n/2}$ and $\delta_{\text{min}}(t) \sim 2^{-n}$ according to Figure 9. Equation (18) is the mathematical proof that the largest scale in a SR mixer decays at half the rate of the scales of a baker’s transformation, $\Lambda_{\text{theo}} = \ln(2)$. For the smallest lamella, the thickness decays according to baker’s transformation. This is preliminary evidence that the necessary length of
the mixer will be affected by the fact that the flow has a velocity profile developed by the wall effects.

![Graph showing rate of mixing scales decay](image)

*Figure 9 Rate of mixing scales decay for $r_z = 1$ calculated for the thickness of the largest and smallest strips.*

**Design of SR Mixers from Striation Thickness Distribution**

SR mixers have typically small dimensions, and generally, in these devices, convection is much faster than molecular diffusion. The rate of segregation scales decay plays an important role in the complete mixing at the molecular scale. The mixing quality can be calculated from the intensity of segregation at molecular scale, which was defined by Danckwerts (1952) as

$$I_s(t) = \frac{\sigma_C^2}{\bar{C}(1 - \bar{C})}$$

(19)

where $t$ is the time, $\sigma_C^2$ is the variance of the concentration of species 1, and $\bar{C}$ is the mean concentration.

The quality of mixing should be analysed from the intensity of segregation determined from the worst-case scenario, i.e. for the mixing of the two largest strips, and for $r_z = 1$. For these conditions, the description of concentration over space and time may be determined from the
lamellar model described by Ottino et al. (1979). In this model, the coordinate system is set up at the interface of the two lamellae, and the conservation equation is given by

\[ \frac{\partial C}{\partial \tau} = \frac{\partial^2 C}{\partial \xi^2} \]  

(20)

where \( \xi = x/s(t) \) is a non-dimensional space scale based on the instantaneous value of the striation thickness, \( s(t) \), and \( \tau \) is the warped time given by

\[ \tau = \frac{1}{t_D} \int_0^\tau \eta(t')^2 dt' \]  

(21)

where \( \eta(t') = s_0/s(t') \) and \( t_D = s_0^2/D_m \) is the characteristic diffusion time for an initial striation scale \( s_0 \), and molecular diffusivity \( D_m \) (Ottino et al., 1979). Note that, by definition, \( s(t) = (\delta_1 + \delta_2)/2 \), where \( \delta_1 \) and \( \delta_2 \) are the thickness of each strip (Ottino et al., 1979). The worst-case scenario studied in this work considers the mixing of the two largest strips, \( \delta_1 = \delta_2 = \delta_{\text{max}} \), and so \( s(t) = \delta_{\text{max}}(t) \).

For the specific case of SR mixers, the time \( t' \) in Equation (21) can be written as a function of the number of elements,

\[ t' = n \frac{V_{\text{element}}}{q} \]  

(22)

where \( V_{\text{element}} \) is the volume of each mixing element, \( V_{\text{element}} = NL^3 \), wherein \( N \) is the geometrical parameter, and \( q \) is the volumetric flow rate, \( q = L^2v \). Equation (22) can be replaced in Equation (21) and

\[ \tau = \frac{2N}{Pe} \int_0^n \delta(n')^{-2} dn' \]  

(23)

In Equation (23), Peclet number is given by \( Pe = s_0v/D_m \) and \( \eta(n) = \delta_{\text{max}}^{-1}(n) \) for two large lamellae. Note that, for the sake of simplicity, it has been considered that the variable \( n \) is continuous.

Equation (20) describes the changes of species concentration in the normal direction of the interface when a lamellar structure is formed by the impingement of reactants in SR mixer.
Considering that two fluids are injected through two opposed jets, the initial condition of Equation (20) is given by

\[
\tau = 0; \ C = H(\xi), \forall \xi \in \left[-\frac{1}{2}, \frac{1}{2}\right]
\]  

(24)

where \(H(\xi)\) is the Heaviside function. The boundary conditions correspond to the gradient of concentrations at the boundaries of a single stria, \(\xi = -0.5\) and \(\xi = 0.5\), which is equal to 0,

\[
\tau > 0; \ \frac{\partial C}{\partial \xi} = 0 \text{ for } \xi = \pm \frac{1}{2}
\]  

(25)

The PDE equation in Equation (20) has the following analytical solution

\[
C(\xi, \tau) = \frac{1}{2} \left( 1 + 4 \sum_{n=1}^{\infty} \frac{1}{\pi n} \sin(n\pi \xi) \exp(-n^2 \tau) \right)
\]  

(26)

Equation (26) describes the evolution of the diffusive front thickness in a mixing element for a non-dimensional space scale and warped time. This expression is used to calculate the intensity of segregation (Equation (19)),

\[
I_s(\tau) = \frac{\int_{-1/2}^{1/2} (C(\xi, \tau) - \bar{C}(\tau))^2 d\xi}{\bar{C}(\tau)(1 - \bar{C}(\tau))}
\]  

(27)

where \(\sigma_{\tau}^2 = \int_{-1/2}^{1/2} (C(\xi, \tau) - C(\xi, \tau))^2 d\xi\) and \(\bar{C}(\tau)\) is

\[
\bar{C}(\tau) = \int_{-1/2}^{1/2} C(\xi, \tau) d\xi
\]  

(28)

The intensity of segregation can take values from 0 to 1. For \(I_s = 0\), it means that the system is completely homogenised, \(C(\Gamma) = \bar{C}(\tau)\), and for \(I_s = 1\), fluids are completely segregated.

Figure 10a shows the evolution of the intensity of segregation as a function of \(\tau/2\), determined from Equation (27). The worst-case scenario was considered for the determination of the intensity of segregation, i.e. \(\delta(n) = \delta_{\text{max}}(n)\) in Equation (12). As the warped time increases \((0 < \tau/2 < 1)\), \(I_s\) decreases sharply in an exponential decay. This evidences once again that the striation thickness decay contributes to the reduction of the concentration gradients,
enhancing the molecular diffusion. In this case, the design criterion was $\delta_{\text{max}}(n)$, although one could also opt for the weighted calculation of $I_s$ through any section in the mixer, since the distribution of thicknesses is available from Equation (9).

Figure 10b shows the evolution of $\ln (\tau \text{Pe}/(2N))$ as a function of the number of elements $n$, determined from Equation (23). The Peclet number value is given by $\text{Pe} = \text{Re} \times \text{Sc} = 1 \times 10^9$, which was determined considering a Reynolds number where the flow can already be considered in Stokes regime ($\text{Re}=0.1$) and a Schmidt number of $\text{Sc} \sim 1 \times 10^{10}$, which is a typical value for very viscous liquids. The number of cubic elements considered was $N = 22$. According to Equation (23), the term $\ln (\tau \text{Pe}/(2N))$ is calculated from $\int_0^n \delta(n')^{-2} dn'$. This integral can be determined taking into account the theoretical expression $\delta_{\text{theo}}(n) = 2^{-(n+1)}$ or the model for the thickness of the maximum mixing scale described in Equation (12). The analysis of these two curves shows the differences between $\delta_{\text{theo}}$ and $\delta_{\text{max}}$. A complete description of the striation thickness decay in SR mixers is crucial for a good estimation of the mixing quality.

The impact of striation thickness decay in diffusion may be identified from these results. In a SR mixer, the thickness of the largest lamellae fully controls the efficiency of mixing at molecular scales. These scales are thicker than the ideal ones, and so the design of mixers must be adapted in order to achieve the same mixing degree. Figure 10 gives useful tools for the prediction of the reactor length required to achieve the complete mixedness. For $I_s = 10^{-3}$ that corresponds to 99% of the mixing efficiency, the mixer length must have, at least, 11 elements for an SR mixer, if the mixing mechanisms are described from the ideal baker’s transformation. However, the same intensity of segregation is achieved in SR mixers with 20 elements. This evidences that the length of a SR mixer has to be 1.81 times the length of the idealised reactor to achieve complete mixedness. In terms of design, the reactor must be twice longer than the ideal mixer.
In sum, the validation of the decay of the largest lamellae from CFD simulations enables to use of this model as a design tool in SR mixers. The number of elements required to achieve the same mixing degree predicted from ideal flow model in SR mixers can be determined from the description of the maximum mixing scales decay in the Stokes regime.

![Graph](image)

**Figure 10** (a) Intensity of segregation as a function of a warped time $\tau$ and (b) $\ln(\tau Pe/(2N))$ as a function of a number of elements $n$.

**Conclusions**

Mixing highly viscous liquids in non-inertial flows is a challenging task due to the absence of turbulent eddies or chaotic dynamic flow structures that promote efficient mixing. To this end, the technology studied here is mixing in Split-and-Recombine mixers, which ensures the generation of multiple fluid lamellae even at vanishingly low Reynolds numbers. CFD simulations results show that the thickness of fluid striations, developed in split-and-recombine mixers, are not uniform in the Stokes regime. An analytical expression, which describes the thickness distribution of scales for any flow rate ratio, is proposed and validated. The impact of assuming an ideal striation thickness decay on the complete mixing at the molecular scale of the full domain was also addressed.

These results give an explicit and straightforward design expression to calculate the maximum and minimum striation thickness of each phase, that correspond to the limits of mixing.
performance in this type of mixers. The validation of this expression shows that the maximum striation thickness decays at half the rate of the thickness described by the ideal scenario given from the baker’s transformation, according to the Lyapunov exponent. This conclusion was the first evidence that the reactor is affected by the shearing profile of the real flow in addition to the prevalent folding mechanisms.

The study on the intensity of segregation of the two largest strips considers the worst-case scenario because, for this condition, the mixing degree is controlled at the molecular scale. Results show that complete mixedness is achieved in a reactor with 1.81 longer the device tailored from the baker’s transformation. The design methodology proposed in this work is more industrially relevant than the tools previously presented in the literature, i.e. the average Lyapunov exponent.

The operation conditions set for the SR mixer in this work are Re ≪ 1. If the mixing device is operated at laminar inertial flow regimes rather than in the Stokes regime, the striation thickness decay would be faster due to flow engulfment, and thus the present work is not applicable to such regimes. In the Stokes regime, the definition of the number of elements is the only parameter setting the performance of split-and-recombine mixers in the industry. The development and validation of a model that predicts the scale and intensity of segregation in SR mixers at Stokes flow regime is a valuable design tool for industrial applications.

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**Notation**

- $a$ – parameter to define if the mixing element is odd or even
- $A$ – area [m$^2$]
- $C$ – concentration [kg m$^{-3}$]
- $\bar{C}$ – mean concentration [kg m$^{-3}$]
- $D_m$ – diffusional coefficient [m s$^{-2}$]
- $h$ - height of the square duct [m]
- $I_s$ – intensity of segregation
- $l$ - length of the square duct [m]
- $L$ – unit of length
- $n$ – number of mixing elements
- $n$ – normal direction
- $N$ – number of cubic elements
- $p$ – pressure [Pa]

Pe – Peclet number
\( q \) – volumetric flow rate

\( Q \) – volume of flow rate

\( r_s \) – flow rate ratio

\( \text{Re} \) – Reynolds number

\( s(t) \) – striation thickness \([\text{m}]\)

\( s_0 \) – initial striation thickness \([\text{m}]\)

\( \text{Sc} \) – Schmidt number

\( t \) – time \([\text{s}]\)

\( t' \) – time \([\text{s}]\)

\( t_D \) – instantaneous diffusional time \([\text{s}]\)

\( u \) – velocity

\( V_{\text{element}} \) – volume of each cubic element \([\text{m}^3]\)

\( X \) – initial position

**Greek Letters**

\( \beta_c \) - aspect ratio of the channel given by the ratio between height and length of the cross-section channel

\( \delta \) – thickness of each strip

\( \delta_{\text{max}} \) – thickness of the largest strip

\( \delta_{\text{min}} \) – thickness of the smallest strip

\( \varepsilon \) – position of each interface

\( \eta \) – area stretch
\( \lambda \) – stretching function

\( \Lambda \) – Lyapunov exponent

\( \mu \) – viscosity [Pa·s]

\( \xi \) – space scale based on the instantaneous value of the striation thickness

\( \xi^* \) - non-dimensional spatial coordinate based on the length of the square duct

\( \rho \) – density [kg m\(^{-3}\)]

\( \sigma_c \) – variance of concentration

\( \tau \) – warped time [s]

\( \nabla \) – velocity [m s\(^{-1}\)]

\( \phi_1 \) – volume fraction of fluid 1

\( \psi^* \) - non-dimensional spatial coordinate based on the height of the square duct

**Subscripts**

1 – fluid 1

2 – fluid 2

\( i \) - discrete transform variables in the sine Fourier transform

\( j \) - discrete transform variables in the sine Fourier transform

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