TRANSPORT PHENOMENA IN PASSIVELY MANIPULATED CHAOTIC FLOWS: SPLIT-AND-RECOMBINE REACTORS

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ABSTRACT
Static mixers and multifunctional heat exchangers/reactors are being used increasingly in process industries. In the inertial or turbulent regime, mixers often incorporate inserts or corrugated walls whose primary function is to create embedded flow vorticity. On the other hand, in low-Reynolds-number flows, for viscosity or residence time purposes, it is necessary to provide solutions based on kinematic mixing, i.e. the topology of the primary flow, such as split-and-recombine reactors (SAR). The concept is based on passive liquid stream division, then rotation in bends of opposite chiralities, and finally recombination, achieving stretching/folding following the baker’s transform. Mixing is efficiently ensured by diffusion without generating prohibitive pressure drops. In this work, a chemical probe is used to study mixing and mass transfer in two different split-and-recombine square duct geometries, SAR-1 and SAR-2 of 3 mm side. Results show that effective mass transfer and mixing can be achieved with a short reactor length and moderate pressure losses; the SAR-1 geometry being more efficient. The chaotic configurations are a good compromise even for higher Reynolds numbers compared to static mixers operating in the transitional regime: they produce moderate pressure losses while enhancing mass transfer.

KEYWORDS
Mass transfer, chaotic advection, split-and-recombine reactor, mixing in industrial processes, iodide-iodate chemical probe

INTRODUCTION
Multifunctional heat exchangers/reactors are containers for thermally active chemical synthesis processes with demanding safety and efficiency requirements. These processes require good mixing properties and must avoid irreversible structure changes in fragile fluid products such as in the pharmaceutical industry. Food processing, for example, often involves highly viscous fluids, many of which are also non-Newtonian [1]. In addition, a primary challenge in this type of technology is increasing the heat removal (or supply) in the system while

NOMENCLATURE

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>A</td>
<td>light absorbance</td>
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<tr>
<td>d</td>
<td>H⁺ injection needle diameter (m)</td>
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<tr>
<td>D_h</td>
<td>channel hydraulic diameter (m)</td>
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<tr>
<td>L₀</td>
<td>initial characteristic dimension of the interfacial area</td>
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<tr>
<td>L(t)</td>
<td>characteristic dimension of the interfacial area at time t</td>
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<tr>
<td>l</td>
<td>length of one SAR unit (m)</td>
</tr>
<tr>
<td>n</td>
<td>number of splitting and recombination (SAR) steps</td>
</tr>
<tr>
<td>Pe</td>
<td>Péclet number</td>
</tr>
<tr>
<td>Q_H⁺</td>
<td>flow rate of injected sulfuric acid (m³ s⁻¹)</td>
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<tr>
<td>Q_p</td>
<td>flow rate of main flow (m³ s⁻¹)</td>
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<tr>
<td>Re</td>
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<tr>
<td>t_mix</td>
<td>mixing time (s)</td>
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<td>U</td>
<td>inlet flow velocity (m s⁻¹)</td>
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<tr>
<td>W</td>
<td>mean flow velocity (m s⁻¹)</td>
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<tr>
<td>X_S</td>
<td>segregation index</td>
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Greek symbols

<table>
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<tr>
<th>Symbol</th>
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<tbody>
<tr>
<td>ε</td>
<td>turbulent kinetic energy dissipation rate (m² s⁻³)</td>
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<td>η</td>
<td>mixing efficiency</td>
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<tr>
<td>λ</td>
<td>interfacial stretching</td>
</tr>
<tr>
<td>ν</td>
<td>kinematic viscosity (m s⁻²)</td>
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<tr>
<td>σ</td>
<td>Lyapunov exponent</td>
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working in the laminar regime to maintain sufficient residence time for reactive chemistry. Mixing, heat transfer, and dispersion can be difficult because of the large energy costs of processing these fluids due to their shear-sensitive complex-structure rheology and possible irreversible denaturation. Reactions of biological agents require gentle yet rapid mixing of reagents as well as meticulous temperature control to inhibit undesirable side reactions [2]. Scaling-up for product quality requires simultaneous consideration of both mixing and heat transfer, so that industrial laminar flow mixers and heat exchangers must be specially designed for these purposes.

The way to intensify the transport phenomena is then either to induce unsteady flows via metallic foams, fins, corrugated channels, wall curvature and the like [3-9] or to create particular three-dimensional structures in the steady flow reaction path, as in the chaotic flux recombination reactors discussed here and shown in Fig. 1. The configuration names SAR-1 and SAR-2 reflect the number of splits and recombinations in the repetitive elementary unit. They were first proposed on a microscale by Gray et al. [10] and Chen and Meiners [11] respectively. The network of separated and then recombined channels in these configurations creates chaotic structures while maintaining the flow in the laminar regime. They present topological structures that exploit the flow laminarity to fold the flow repeatedly and double the lateral striation. This creates a layered opposite directions, and recombined, folding the concentration of baker’s transforms on the concentration profile. The two layer per microfluidic element, this topology performs a series of reactor, the subject of this study, a re then discussed here and shown in Fig. 1. The configuratio

The baker’s map, the mathematical background for mixing by cutting and stacking, is discussed in the section that follows. The principle and manufacturing technique of the flux recombination reactor, the subject of this study, are then presented. The method for quantifying mixing of the chemical probe is then briefly introduced. The results are then discussed and compared with available data; the friction losses are also presented. The final section draws some conclusions and discusses the use of such devices in process industries.

FIG. 1. SPLIT-AND-RECOMBINE MIXER CONFIGURATION ELEMENTARY UNITS (a) SAR-1 AND (b) SAR-2

FLUX RECOMBINATION REACTORS

The baker’s map

The baker’s map is a mathematical transformation used in conservative dynamical systems [12-14]. The baker’s map is obtained by iterating the transformation \( S \) defined on the unit square by:

\[
S(x, y) = \begin{cases} 
(2x, y) \mod 1, & \text{if } 0 \leq x < \frac{1}{2} \\
(2x, (y + 1) \mod 1) \mod 1, & \text{if } \frac{1}{2} \leq x < 1
\end{cases}
\]  

(1)

The upper and lower halves of the unit square are mapped when the transformation \( S \) is applied repeatedly. As sketched in Fig. 2, squeezing in the \( y \) direction while stretching in the \( x \) direction, cutting in two and then stacking the right half above the left, causes what is initially a two-strip domain to become \( 2^{n+1} \) strips after \( n \) iterations. The baker’s map is a mixing transformation since, for a sufficiently large number of iterations (limit \( n \to \infty \)), the unit square becomes completely filled with an infinite number of alternating red and blue lines of decreasing thickness after each iteration, and thus any infinitesimal region of the square contains the same proportion of the two colors.

To reproduce this transformation in real mixers, the fluid flow must encounter spatial discontinuities. Flux separation/recombination reactors have three-dimensional stationary continuous mixer configurations intended to mimic the transformation \( S_n \). For instance, a basic element of the SAR-1 configuration is composed of a converging zone followed by a diverging zone at right angles; the two outlets of the diverging zone are connected to the converging zone of the next element by two opposite ducts. The converging zone plays the role of the “stretching/squeezing” step while the diverging one is

FIG. 2. THREE ITERATIONS OF THE BAKER’S TRANSFORM APPLIED TO THE UNIT SQUARE
the “cutting” step. In the outer ducts, red and blue regions are inverted, ensuring the “stacking” step. If red and blue fluids entering the mixer form a two-layered domain in the first element, they should form a four-layered domain at the entry of the second, just as in the baker’s map. The same reasoning applied to the second element leads to eight strips at the entrance of the third element, and so on. If the segregation scale at the reactor inlet is \( l_0 = h \), where \( h \) is the characteristic channel length, after \( n \) elements \( l_n = h/2^n \).

**Chaotic advection**

As described above, split-and-recombine mixing relies on a multistep procedure. The basic operations are: splitting of a bi- or multilayered stream in the transverse direction and recombination of the resulting substreams. One or more reshaping steps usually accomplish these basic flow manipulations. From the perspective of chaotic advection, the mixing performance is commonly characterized by the interfacial stretching, \( \lambda \):

\[
\lambda(t) = \lim_{t \to t_0} \frac{L(t)}{L_0}
\]

where \( L_0 \) and \( L(t) \) denote the characteristic dimensions of the lamellar interfacial area at \( t = 0 \) and at finite time \( t \). In 2D laminar incompressible flow, the diffusive mass transport, which determines the mixing performance, depends quadratically on the interfacial stretching. Chaotic flows enhance mixing by exponentially increasing stretching over time. Accordingly, the finite-time Lyapunov exponent \( \sigma \) may be defined by Eq. (3):

\[
\lambda(t) \approx e^{\sigma t}
\]

The lamellar pattern remains highly regular and well defined after repeated application of the SAR principle. However, chaotic behavior is exhibited in the exponential increase of the interfacial area between these lamellae. The corresponding stretching factor can be expressed by:

\[
\lambda(t) = (2^n - 1) = 2^{\text{SAR}/t}
\]

where \( n \) denotes the number of SAR steps, \( W \) the mean velocity, and \( l \) the length of one SAR element. According to Eqs. (3) and (4), the SAR mixer has a finite-time Lyapunov exponent \( \sigma = \ln 2 \). Thus, even if the flow pattern is highly regular, chaotic advection can be achieved in these configurations, as reflected in the positive Lyapunov exponent [15]. In addition to the regions of high chaos where mixing predominantly occurs, chaotic advection generally induces regions of regular flows exhibiting poor mixing. However, in the case of an ideal SAR multilamination, a spatially homogeneous mixing is obtained after a reasonable number of cutting and stacking steps, as shown in Fig. 3. Since the final lamella dimension depends on the number of SAR steps and not only on the channel width, efficient mixing can be achieved under moderate pressure drops. The pressure drop achieved remains linearly correlated to the number of SAR units while the lamella dimension decreases exponentially with each step.

The present work, however, focuses on the experimental characterization of mixing qualities in the SAR-1 and SAR-2 configurations using the chemical probe method. This technique ultimately allows quantitative determination of the mixing rate and classification of the two multifunctional heat exchangers/reactors based on their mixing performance. The present method does not give access to the chaotic nature of the flow, nor to the calculation of the corresponding Lyapunov exponent.

Several similar flow SAR reactors have been studied. Schönfeld et al. [16] have introduced an optimized split-and-recombine micromixer with uniform chaotic mixing. A chaotic micromixer using two-layer crossing channels to exhibit fast mixing at low Reynolds numbers was studied by Xia et al. [17]. Ohkawa et al. [18] investigated flow and mixing characteristics in what they called a \( \sigma \)-type plate static mixer with splitting and inverse recombination.

**FIG. 3. SIMULATIONS OF ASSOCIATED MIXING IN (a) SAR-1 AND (b) SAR-2 [3]**

**TEST SECTION DESIGN AND FABRICATION**

To fabricate the microscale SAR-1 configuration, Gray et al. [10] used a combination of silicon fusion bonding and deep reactive ion etching. Chen and Meiners [11] used a standard planar multilayer soft lithographic technique for SAR-2.

In the present work, these configurations are reproduced in 316L stainless steel on a mini-scale in order to assess their potential use as mixers and/or heat exchangers in the process industry. Stacking multiple plates specially designed to constitute the required flow path when assembled is the most common technique. The technological solutions for the manufacturing process include powder compaction by high-temperature compression, laser fusion of metal powder, and replicative processes such as casting and welding/diffusion by high temperature isostatic compression. This last technique includes putting in contact, at high temperatures and pressure for a period of time, two plates of a given material. Plates are welded by diffusion of atoms so as to form a connecting structure similar to the basic structure of the material. The
welding procedure has two cycles: a first low-pressure cycle where the plates are held at a temperature between 1000 to 1100 °C and a pressure between 50 and 150 bars for 2 hours, and a second high-pressure cycle where the temperature remains around 1000 °C and the pressure is increased to around 1000 to 1500 bars for another 2 hours. The technique is relatively simple to implement and the plates can be machined in advance by laser cutting, which has the advantages of dimensional accuracy, control, and reliability. Figure 4 illustrates the manufacturing process and the design of the six plates constituting the core of a variant of the Chen and Meiners reactor configuration. The mockups were fabricated at the Commissariat à l’Énergie Atomique et aux Énergies Alternatives – CEA laboratories, Grenoble, France; a patent on the manufacturing technique was recently filed [3,19].

FIG. 4. ELEMENTARY PLATES ILLUSTRATING THE TECHNIQUE FOR MOCKUP FABRICATION [19]

The channels have a 3×3 mm² square cross section with hydraulic diameter \( D_h = 3 \text{mm} \). SAR-1 is made of 48 elements distributed over four parallel rows with a total developed length (sum of the lengths of each branch of the 3D channel) of 2669 mm. SAR-2 includes 7 rows of 6 elements each, with a total developed length of 3134 mm. The two mixers are designed to exhibit comparable residence times for the range of fluid velocities studied. The Reynolds number (Eq. (5)) is calculated at the mixer inlet for the maximum flow velocity with water being the working fluid:

\[
Re = \frac{UD_h}{v}
\]  

(5)

To assess the evolution of mixing properties with the flow regime, \( Re \) is varied between 40 and 5000 corresponding to water mass flow rates between 0.43 and 54 kg h⁻¹. In this range, the SAR reactors can be used in production lines to process fluid material on a scale larger than that of microfluidics (to which their applications are currently still limited).

MIXING QUANTIFICATION BY THE CHEMICAL PROBE METHOD

The iodide-iodate reaction system

The essentials of the chemical probe method are briefly presented in this section. For further information and detailed discussion the reader can refer to Baldyga and Bourne [20,21]; Fournier et al. [22]; Guichardon and Falk [23]; Falk and Commenges [24]; Durandal et al. [25]. The adaptive procedure of the chemical probe method, as described by Hachbi et al. [26] and Ghanem et al. [27], is adopted in the present study. This procedure consists of fitting the concentrations of the injected sulfuric acid to each flow condition in order to control the reaction volume and increase the accuracy and relevance of the results.

The iodide-iodate system is based on competitive parallel reactions: the quasi-instantaneous borate neutralization (Eq. (6)) and the Dushman reaction (Eq. (7)), which is much slower. The balanced reactions can be modeled as follows:

\[
\text{H}_2\text{BO}_3^- + \text{H}^+ \rightarrow \text{H}_3\text{BO}_3^- \quad (6)
\]

\[
5\text{I}^- + \text{IO}_3^- + 6\text{H}^+ \rightarrow 3\text{I}_2 + 3\text{H}_2\text{O} \quad (7)
\]

The iodine \( \text{I}_2 \) further reacts with iodide ions \( \text{I}^- \), yielding \( \text{I}_3^- \) ions following the quasi-instantaneous equilibrium reaction:

\[
\text{I}^- + \text{I}_2 \leftrightarrow \text{I}_3^- \quad (8)
\]

Figure 5 illustrates schematically the method whose principle is to add, in stoichiometric deficit, a small quantity of sulfuric acid \( \text{H}^+ \) to an initial mixture of \( \text{I}^- \), \( \text{IO}_3^- \), and \( \text{H}_3\text{BO}_3^- \). In perfect mixing, the injected \( \text{H}^+ \) is totally consumed by the quasi-instantaneous reaction in Eq. (6) and hence there is no formation of iodine \( \text{I}_2 \). When the mixing process is not fast enough to sustain the first reaction, the local overconcentration of \( \text{H}^+ \) produces iodine \( \text{I}_2 \) by the second reaction (Eq. (7)), which reacts with iodide ions \( \text{I}^- \) to yield \( \text{I}_3^- \) ions (Eq. (8)); this modifies the absorbance of the final solution, which is detected by the spectrometer to deduce the concentration of produced \( \text{I}_3^- \). Thus the selectivity in \( \text{I}_3^- \) is a measure of molecular-scale segregation and indicates mixing quality. The characteristic segregation index \( X_s \) is defined by the expression:

\[
X_s = \frac{Y}{Y_{ST}}
\]

(9)

where \( Y \) is the ratio of the quantity of \( \text{H}^+ \) transformed into \( \text{I}_2 \) following the reaction in Eq. (7) to the total quantity of injected \( \text{H}^+ \), and \( Y_{ST} \) is the value of \( Y \) in the case of total segregation. \( X_s \) is given by Villermaux [28] for open loop flows as:
\[ X_S = \frac{2([I_2] + [I_I])}{[H^+ + 1]} \left( \frac{Q_{in}}{Q_{out}} \right) \left( \frac{H_2BO_4^-}{6[IO_3^- + 1]} \right) \]  

where the slower Dushman reaction dominates the quasi-towards unity, we speak of total segregation, a state of poor mixing where the slower Dushman reaction dominates the quasi-time instantaneously borate neutralization. Similarly, values of the segregation index around zero indicate perfect mixing qualities.

The quantitative mixing time \( t_m \) can then be linked to \( X_S \) using physical models detailed in the references mentioned above; we present here the main equations for the models utilized for the different flow regimes.

Theoretically, the stretching efficiency model proposed by Falk and Commenge [24] is valid for the laminar regime where shear rates, stretching, striation thickness, and deformation tensors are examined and mixing time is consequently calculated as:

\[ t_m = \frac{D_k}{8W\eta} \arcsinh \left[ 3.04 \left( \frac{d}{D_k} \right)^2 Pe\eta \right] \]  

In the turbulent regime, following the models of Baldyga and Bourne [20, 21], \( t_m \) is proportional to \( \varepsilon^{1/2} \), and by dimensional analysis, we can ultimately deduce that \( t_m \) is proportional to \( W^{-3/2} \) and thus to \( Re^{3/2} \).

On the experimental side, to pass from the qualitative case-specific segregation index to global mixing quantification, the engulfment model (E-model) formulated by Baldyga and Bourne [20] is used. This model is based on the hypothesis that the viscous-convective process controls mixing at scales comparable to the Kolmogorov scale. Thus, the engulfment rate \( E \), the inverse of the mixing time, is related to the TKE dissipation rate \( \varepsilon \) and the kinematic viscosity \( \nu \) following the scale-law analysis by Baldyga and Bourne [20] by considering the rate of growth of the volume \( V_i \) containing the species \( i \):

\[ \frac{dV_i}{dt} = EV_i \]  

\[ E = \frac{1}{t_m} = 0.058 \left( \frac{\varepsilon}{\nu} \right)^{3/2} \]  

When Eq. (12) is expressed in terms of mass balance, the growth of the uniform mixing zone of concentration \( c_i \) is given by the temporal differential equation:

\[ \frac{dc_i}{dt} = E(c_i > -c_i) + R_i \]  

where \( < c_i > \) is the mean concentration value of species \( i \) in the environment when mixing is completed and \( R_i \) is the rate of formation of substance \( i \) by the chemical reaction. The calibration curves for the chemical probe are obtained by solving Eq. (14) for each of the involved species using the Newton–Raphson iterative method [26,27]. Ultimately, the segregation index is linked to the engulfment rate whose inverse gives the value of \( t_m \).

**EXPERIMENTAL PROCEDURE**

The initial reagents are in the solid phase, with purity around 99%. Their mass is measured by an analytical balance of 5×10⁻⁵ g precision. They are then dissolved in deionized water (on a Siemens resin < 5 µm) and stored in the main tank. The resulting solution is mixed by an immersed pump and its temperature is kept constant by a thermostatic bath at 298 K. Another important parameter considered in dosing the reactants is the pH. The acidity of the medium should be controlled so as be high enough to prohibit the natural formation of iodine in the absence of acid molecules, but not so high as to destabilize the iodine dissociation after the reactions are completed. The ideal initial pH value was found to be between 8.5 and 9.5. For more details on the pH-potential diagram, see Guichardon and Falk [23].

The mixture is driven by a variable-speed rotary pump into the hydraulic loop. The main flow rate is measured by a series of rotameters depending on the flow rate with a precision of 2%. The acid is injected at the entry of the channel system by a needle of 0.5 mm internal diameter and its concentrations are adapted to ensure global measurements over the entire reactor length. The injection setup consists of a regulated stepper motor connected to a push-syringe system. The flow rate of the injected sulfuric acid could influence the results since it can perturb the main flow. Therefore a study was made to determine the maximum flow rate of the injected sulfuric acid for which there is no influence on the measured values. It was found that when \( Q_a/Q_H^+ > 200 \), there is no effect on the...
Segregation index, and hence this ratio is fixed for all the measurements. The acid injection flow rates are controlled by a velocity regulator on the angular velocity of the stepper motor via the software IMS terminal through a PC.

The reactor is preceded by a straight-pipe preconditioner of length 2 m to ensure fully developed flow at the reactor inlet, and is followed by a postconditioner of length 0.3 m, after which a part of the flow is directed towards the cistern of the spectrophotometer in order to measure its absorbance A and thus calculate the segregation index. The I$_2$ and I$_3^-$ concentrations are experimentally determined by spectrophotometry. According to the Beer-Lambert law in Eq. (15), the light absorption A is proportional to the I$_3^-$ concentration resulting from the equilibrium reaction (Eq. (8)):

$$[I_3^-] = \frac{A}{\xi \ell}$$

where $\ell$ is the optical length and $\xi$ is the molar extinction coefficient of I$_3^-$ at 353 nm: $\xi = 2597 \pm 148$ m$^2$/mol (Palmer et al. [29]). Once $[I_3^-]$ is measured, the concentration of I$_3^-$ can be obtained from the expression for the mass balance of iodine. The final products of the chemical reaction system are continuously analyzed through a channel placed 0.3 m downstream from the reactor outlet to a quartz recirculation cistern of capacity 1 mL. The spectrometer (Jenway 6505™) wavelength range is in the ultraviolet domain [190 nm, 1100 nm] and resolution is 0.1 nm with a bandwidth of 1.8 nm. The measurable absorbance range lies between 0 and 3 with 0.1% precision.

RESULTS

The experimental measurements of the tri-iodide ion concentration in each of the reactor configurations studied ultimately allow their characterization. Their mixing qualities are presented and compared in terms of segregation index, mixing time, and inverse diffusion coefficient. Energy expenditures are conveyed by the experimental absolute pressure drop and the dimensionless friction factor.

Segregation index $X_S$

The segregation index is the signature of the micromixing process. It gives a clear idea of the evolution of mixing enhancement with flow hydrodynamics, namely the Reynolds number. The segregation index $X_S$ is a criterion to differentiate and compare different flow behaviors when measured under the same operating conditions and reagent concentrations. Different concentrations of H$^+$ varying between 0.0025 and 0.1 mol L$^{-1}$ are injected for the studied range of main-flow Reynolds number, the H$^+$ concentration being “adapted” for each flow velocity to ensure that the Dushman reaction (Eq. (7)) takes place over a reactive volume that exactly corresponds to the reactor length in order to ensure global mixing-time measurements. Special attention was given to the low-Reynolds-number zone where the flow is deeply laminar or inertial, since the interest of such configurations lies in this zone. The absorbance of the solution at the outlet is continuously measured by the spectrophotometer and the segregation index $X_S$ is thus calculated. Figure 6 plots the evolution of $X_S$ as a function of the Reynolds number for the two mixers.

![FIG. 6. SEGREGATION INDEX VERSUS Re](image)

The effect of increased inlet flow velocity and the resulting turbulent structures on enhancing mixing qualities can be clearly seen. $X_S$ shows a rapid decrease with increasing Reynolds number especially in the laminar regime, implying increased rates of stretching and folding and thus better selectivity and enhanced mixing. Starting from the value of 1000, the effect of increasing Reynolds number on mixing enhancement is less accentuated: $X_S$ seems to stabilize and little enhancement is produced by further increasing Re. This is the threshold at which the contribution of turbulent structures to mixing starts to mask the effect of the chaotic advection mechanism. This draws attention to the fact that such geometries are mostly interesting in laminar regimes. In addition, their use in highly dissipative turbulent flows might drastically increase pumping costs, which remain a central concern in the design and sizing of new devices.

It should be noted that the effect of the geometry remains visible even in the turbulent regime, and that the configuration SAR-1 shows lower levels of segregation over the entire range studied. This can be explained by the fact that the SAR-1 elementary unit imposes more flow direction changes on the fluid stream than SAR-2. The fluid entering the inlet of SAR-1 unit flows through three different superposed planes, while in SAR-2 it is restricted to two. Naturally, more direction changes are needed to return to the starting level at the following
element entry; this active circulation at the elbows accentuates mass transfer and additional flow energy is dissipated to enhance mixing.

Nevertheless, the segregation index remains qualitative and case-specific. Quantitative mixing times that are independent of experimental and chemical conditions are presented in the following sections as a function of flow regime and energy consumed per unit mass of processed fluid as calculated from the inlet-outlet differential pressure drop.

Mixing time $t_m$

Following the method described above, $t_m$ is calculated in each reactor. Mixing time is normalized by the diffusion time calculated at the reactor inlet. By assuming a molecular diffusion coefficient $D_m = 2.229 \times 10^{-9}$ at 25 °C and a lamellar thickness of 1.5 mm corresponding to half of the channel width, the diffusion time is calculated: $t_d = 979$ s. The dimensionless mixing times are plotted against Reynolds number in Fig. 7 with an accuracy of 10% as calculated from the different experimental errors of the various components of the test rig. The results are compared to those obtained in two other reactor geometries using the same chemical probe method under the same operating conditions; these are the wavy and the zigzag rectangular channels (2x4 mm cross section) of hydraulic diameter 2.67 mm. The wavy channel is made of 90° curved bends of curvature radius $R_c = 10.5$ mm. The zigzag channel is made of 90° curved bends of curvature radius $R_c' = 1.5$ mm. Further details on the corrugated channels can be found in Ghanem et al. [27].

The curve of $t_m$ is expected to follow the same general trend as that of $X_s$: as the Reynolds number increases, the rates of stretching and engulfment increase in the flow, enhancing mass transfer and decreasing the characteristic mixing time. In the lower part of its range, the Reynolds number strongly influences the evolution of mixing time: slight increases in the Reynolds number in the laminar zone are accompanied by remarkable decreases in the mixing time, a phenomenon difficult to achieve in deeply laminar plain channel flows where fluid layers tend to flow parallel to one another with little transverse activity. Flow velocity increases the rate of cutting, stretching, squeezing and stacking of fluid strips, thus promoting mass transfer by diffusion.

The wavy and zigzag channels are designed to operate in the inertial zone, unlike the chaotic configurations that are most efficient for laminar regimes; however, the split-and-recombine reactors continue to show better mixing times even in the transitional and turbulent zones since they combine both effects: inertial forces and secondary flows triggered by changing flow direction through bent structures, together with splitting and recombination of the fluid bulk. This coupling between chaotic advection and basic flow hydrodynamics is the reason for the better mixing qualities exhibited by the SAR configurations.

As shown by the segregation index plot of Fig. 6, the SAR-1 configuration exhibits lower levels of segregation than SAR-2. Passing from the qualitative segregation index to the quantitative mixing time following the models in the literature confirms this superiority. The SAR-1 configuration exhibits smaller mixing times over the range studied. Little distinction can be seen for deeply laminar flows, and the gap widens as $Re$ increases. As expected from the theory of the baker’s transform, from a purely kinematic point of view, the two configurations share similar split-and-recombine behavior and produce comparable cutting and stacking effects. What differentiates one configuration from the other is the number of steps and the external connecting channels between the diverging inlet and the converging outlet zones. As the flow’s kinetic energy increases by increasing the velocity, more of this energy is consumed in the bends and dissipated on molecular scales to enhance mixing. From this perspective, the SAR-1 configuration has a more complex 3D structure than SAR-2 and consequently the incoming fluid is passively manipulated more intensely by the bent geometry because it is forced to change its direction more frequently in the geometry in which linear distances in each SAR unit are shorter.

It is the ratio of flow energy consumed for mixing enhancement to that lost by viscous dissipation that classifies the different configurations in terms of energy efficiency. Accordingly, Figure 8 plots from a process engineering point of view the evolution of mixing time with the specific energy consumed per kilogram of treated fluid in each of the reactors. The specific energy represents pumping costs to process, transform, or produce a unit mass of fluid and is calculated from the overall pressure drop in each mixer. It can be seen that, in the laminar zone, up to the specific energy value of 0.5 J.kg$^{-1}$, mixing time decreases rapidly with increasing energy expenditures.
For higher flow regimes, an equal enhancement of mixing qualities requires more energy consumption. This tendency reflects the fact that the real interest of using such configurations is the deeply laminar regime, where most of the flow energy is consumed in the topologic mixing process following the baker’s map. For higher flow velocities, the small channel dimensions and structural singularities produce increased friction losses that render such devices less practical. An interesting fact is that the SAR-1 configuration that shows better mixing under the same hydrodynamic conditions seems to consume less energy; to produce the same mixing time, SAR-2 consumes more energy. In light of these results, it can be seen that the SAR-1 configuration is more efficient and better exploits the flow energy to produce higher levels of stirring and mass transfer, compared to SAR-2 in which more energy is lost by friction. Mixing efficiency is quantitatively discussed in the following section based on the parameter introduced by Ottino et al. [30] and the concept of inverse diffusion coefficient used by Falk and Commenge [24].

Inverse diffusion coefficient – Mixing efficiency

Since only a part of the mechanical energy consumed is used for mixing, Falk and Commenge [24] use the concept of inverse diffusion coefficient \( t_w/D_h^2 \) and mixing efficiency \( \eta \) proposed by Ottino et al. [30] to define the characteristic mixing time that appears in the stretching efficiency model (Eq. (11)).

In addition, to account for the difference in the hydraulic diameters of the tested reactors by the chemical probe method and the possible influence of this parameter, the inverse diffusion coefficient \( t_w/D_h^2 \) proves useful. The variation of this term as a function of \( Re \) is plotted in Fig. 9. The lower the values of this coefficient, the better the mixing qualities. The analysis based on this criterion also shows the superiority of SAR-1. More important on this plot are the validation of the experimental results against the theoretical models and the calculation of the mixing efficiency \( \eta \). In the laminar regime, following the stretching efficiency model [24], \( t_w \) varies theoretically as \( W^{-1} \) (hence \( Re^{-1} \) in Eq. (11)). On the other hand, in the turbulent regime, \( t_w \) varies as \( W^{-0.52} \) and thus \( Re^{-0.32} \) [20, 21]. These theoretical trends are expressed for both configurations by the fitting curves of Fig. 9, showing the existence of two distinct zones as described by the theoretical approach. As the basic flow hydrodynamics is altered by the presence of singularities, together with the interaction between chaotic and vortical structures in the wake of the sharp bends, the regimes do not follow the classical laminar-turbulent transition as in a plain circular tube. The transition is even difficult to detect by conventional pressure-loss measurements. However, a clear transition around Reynolds number 120 can be seen in the plots of the segregation index, mixing time, and friction factor presented in the following section. The influence of flow instabilities and other physical phenomena on this transition are still to be investigated in order to better understand the changes in the flow as it passes from the deeply laminar, creeping state to a relatively inertial regime.

Throughout this work, experimental uncertainties for any derived quantity have been evaluated using the absolute measurement errors for each of its elementary terms. For the inverse diffusion coefficient, the percentage error was estimated at 10% over the range of Reynolds numbers studied. It should be also noted that all experimental data lie within the 30% accuracy interval characteristic of the chemical probe method, as set by Falk and Commenge [24].

The mixing efficiency is deduced by adjusting the values of \( \eta \) in Eq. (11) so that the \( Re^{-2} \) laminar fitting curve matches the experimental values and joins the other curve at the hypothetical transition point. For the SAR-1 configuration, the mixing efficiency is \( \eta = 0.25 \), i.e. 25% of the shear rate is used for mixing; the rest is viscous dissipation. Similarly, an efficiency of \( \eta = 0.15 \) is calculated for SAR-2: a smaller proportion of the dissipated energy is employed to intensify mixing. These results reflect the fact that SAR-1 simultaneously exhibits better mixing times and lower energy consumption: the SAR-1 configuration is more efficient for mixing. Independent of mixing time, an energy efficiency analysis of both configurations is presented in the following section in terms of head losses per unit length and a friction factor.

**FRICTION LOSSES**

The pressure drop is measured by a differential pressure sensor fixed between the reactor inlet and outlet, with water as the working fluid. Figure 10 shows the evolution of head losses as a function of flow rate. The results for the wavy and zigzag channels and those for a plain pipe of equivalent length and diameter are also presented.
The dependence of head losses on the velocity squared is conveyed in the parabolic trends of this plot. The energy consumption in the SAR reactors increases rapidly with $Re$. These devices are especially interesting in laminar flow regimes, where the chaotic mechanisms enhance mass and heat transfer while producing moderate pressure losses. For low flow rates, the curves for the chaotic configurations seem to collapse together as they produce similar head losses. For higher flow rates and flow velocities, major losses in SAR-2 exceed those in SAR-1 and account for the higher overall energy consumption. Naturally, in this type of flow, there are zones in which the fluid flows at a maximum velocity $U$ after being recombined and zones in which the flow is split and the velocity drops to $U/2$. In SAR-2, the ratio of the channel length in which the flow velocity is $U$ to the total reactor length is higher than its equivalent in SAR-1, and as the major losses are proportional to the square of the velocity, this fact accounts for the increased head losses in SAR-2 with little contribution to the mixing process. Furthermore, SAR-2 has a superior developed length, the fluid undergoes more recombinations, and the impacts between the fluid streams in each converging zone produce additional losses.

Surprisingly, both the chaotic geometries produce lower head losses per unit length than the zigzag channel of comparable diameter, although this latter shows poorer mixing qualities in the inertial zone. These additional losses can be explained by the strong fluid impact and recirculation regions in the wake of its sharp bends. Therefore, these configurations can compete with devices operating in the inertial zone since their head losses remain moderate for these flow rates and their use is not limited to laminar regimes.

To isolate the effects of geometric properties, the dimensionless major head loss coefficient $\lambda$ for the chaotic configurations is plotted against $Re$ in Fig. 11. The logarithmic plot follows the same trend as for the plain tube: a linear sharp decrease for low $Re$ ($\lambda \sim 64/Re$), a slight jump in the transitional zone, and a slow decrease for higher $Re$. However, the values of $Re$ marking the boundaries of these zones are different from those found in a plain tube. Flow instabilities induced by the sharp bends certainly promote vortical structures, but further investigation is needed to understand the premature turbulent-like behavior in these configurations.

**CONCLUSIONS**

Mixing in chaotic flux recombination reactors SAR-1 and SAR-2 is investigated experimentally using an adaptive procedure of the iodide-iodate chemical probe method. The configurations, first proposed by Gray et al. [10] and Chen and Meiners [11] on a microscale, are reproduced on a scale that can be exploited in large-scale industrial processes.

Chaotic structures are created in the flow by the effect of three-dimensional chiralities and splitting and recombination of fluid laminates attempting to mimic the mathematical baker’s transform for surface creation. Through continuous stretching, folding and splitting, reorientation and recombination, chaotic advection is generated and the concentrated fluid streams are gradually subdivided into smaller ones. This greatly enlarges the material contact interface and hence significantly promotes mixing phenomena.
A segregation index is extracted from spectrophotometric measurements and quantitative mixing rates are calculated using models from the literature. Comparison of the results to those for other reactors investigated using the same technique shows the flux recombination geometry to be most efficient in the low-Reynolds-number laminar zone. This type of reactor can be used in the pharmaceutical, cosmetic, chemical, and food industries where handling viscous, fragile, and reactive flows is a real challenge. SAR-1 exhibits superior mixing qualities as reflected in lower segregation levels and shorter mixing times. This is confirmed by analysis of the inverse diffusion coefficient, whose plot shows an early transition to turbulent behavior, and the calculation of a mixing efficiency.

On the basis of this criterion, together with energy consumption and friction factor, the SAR-1 configuration shows advantages over SAR-2 and proves interesting for industrial mixing applications. It shows good performance in the low-Reynolds-number laminar zone. This type of reactor shows the flux recombination geometry to be most efficient in mixing times. This is confirmed by analysis of the inverse diffusion coefficient, whose plot shows an early transition to turbulent behavior, and the calculation of a mixing efficiency.

Its heat-transfer qualities are yet to be investigated.

REFERENCES