Numerical study of active mixing over a dynamic flow field in a T-jets mixer – induction of resonance

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Abstract

The flow field in a 2D T-jets mixer was simulated to study the effect of the pulsation/modulation of the jets flow rate on the dynamics of mixing. Different strategies, frequencies and amplitudes of the opposed jets flow rate modulation were tested. The modulation frequencies were set as multiples of the natural oscillation frequencies of the dynamics flow field. The natural flow frequencies are determined from the unforced flow, i.e. when the jets are not modulated. It is found that out phase modulation of the opposed jets, with frequencies close to the natural frequencies, cause resonance of the flow enhancing the order of the system, which results in a flow field with a well-defined repetitive generation of vortices. Conversely, when the pulsation frequencies were different from the natural frequencies the flow disorder is enhanced, i.e. the vortices evolution throughout the T-jets mixers is less repetitive. The impact of the jets flow rate modulation on the flow field dynamics increases with the modulation amplitude up to the extreme case where it completely drives the dynamics of the system. A design equation for the most energy efficient pulsation of the jets feed streams in opposed jets mixers is proposed.

Keywords: Mixing, Laminar flow, Opposed jets mixers, Active mixing, Resonance

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Introduction

Mixing is the critical step for many industrial processes, particularly when the rate of the chemical reaction is faster than the mixing rate, and the reactions are competitive and/or consecutive reactions, e.g., crystallization and reactive polymerization. For those reactions the history of mixing sets the product properties, namely crystallinity or Particle Size Distribution, PSD, or the mechanical properties of polymers obtained by reactive processes (Kolodziej et al., 1982, Kolodziej et al., 1986 and Schwarzer et al., 2006). Schwarzer et al. (2006) and Pieper et al. (2011) demonstrated that the Reynolds number of the jets in a T-jets precipitator has direct impact on the PSD of barium sulphate nanoparticles. The Reynolds number is associated with mixing in T-jets mixers, and thus mixing is an important design parameter for the development of specific products.

The particular case of opposed jets reactors has been often studied for controlled mixing of two liquids, namely for the production of nanoparticles: Marchisio et al. (2006) and Johnson and Prud'homme (2003) used cylindrical mixing chambers with round injectors (Confined Impinging Jets – CIJ) for precipitation of nanoparticles and observed that both the operation Reynolds number and the geometry of the CIJs influence the nanoparticles PSD. Liu and Fox (2006) used CIJs and observed that the conversion of a second order competitive reaction varied three orders of magnitude when the Reynolds numbers at the jets ranged between 1 and 5000. Nunes et al., 2012 also observed the critical role of mixing in a CIJ, reporting the selectivity of a second order competitive reaction that varied three fold over a range of Reynolds numbers less than 300. Schwarzer et al. (2006), Gradl et al. (2006) and Gradl and Peukert (2009) used a prismatic T-jets mixing chamber for the precipitation of barium sulphate; at turbulent or laminar chaotic flow regimes; these authors also observed that the Reynolds number affected the PSD of the nanoparticles. Santos et al. (2005) reported, in a 2D T-jets geometry, that the conversion of a second order chemical reaction depends on the Reynolds number of the jets. Soleymani et al. (2008a) used a micromixing test reaction and varied the reactor geometry and the operation Reynolds number; both factors influenced the chemical reaction conversion. Krupa et al. (2012; Krupa et al. (2014) used a competitive consecutive chemical reaction to assess micromixing in T-jets reactors, and also reported a clear effect of the mixer geometry on the flow regime, which had an impact on the selectivity of the chemical reaction.

Other authors studied mixing in opposed jets mixers, without chemical reaction, using mainly flow or mass transfer data. Engler et al. (2004) studied several T-jets mixers geometries and reported results of mixing obtained from tracer visualization in the range of Reynolds 6 to 200. In this range of Reynolds numbers Engler et al. (2004) identified three flow regimes: stratified flow, vortex flow and engulfment flow. The regimes were characterized as:

- Stratified / segregated flow is a regime where two streams, each fed by one jet, flow without mixing side by side through the mixing chamber. The axis of the mixing chamber defines a symmetry line between both streams. Mixing between the two streams relies solely on diffusion.

- Vortex flow is a regime where there are vortices in the streams flowing side by side without breaking the symmetry line between the streams. At this flow regime, mixing between the two streams still relies solely on diffusion.
- Engulfment flow is a regime where vortices engulf both the streams fed by each jet. Mixing is now mainly promoted by the convective mechanisms – the vortices.

The transition between the flow regimes was not only a function of the Reynolds number; the geometry of the mixers was also influencing the flow regimes transition. Soleymani et al., 2008b also observed that the transition between the three flow regimes was a function of both geometry and Reynolds number. The three flow regimes reported in Engler et al., 2004 and Soleymani et al., 2008b are laminar, even at Reynolds numbers 489, Wong et al., 2004 also reported laminar flow regimes.

In CIJs only two laminar flow regimes are reported: the stratified flow that in CIJ is generally referred to as steady flow regime, and above a transition Reynolds number in the range of 90 to 120 a regime referred in CIJ literature as self-sustainable chaotic flow regime (Lee et al., 1957, Tucker and Suh, 1980, Wood et al., 1991, Johnson and Wood, 2000, Teixeira et al., 2005, Santos et al., 2008). The chaotic flow regime is characterized by oscillations of the jets impingement point and by the formation of vortices immediately downstream the jets impingement point. This flow regime differs from the engulfment flow regime on the orientation of the flow rotation, while in the engulfment flow regime the flow rotation is aligned with the mixing chamber axis, in the chaotic flow regime the vortices rotation is perpendicular to the mixing chamber axis Santos and Sultan (2013). Santos et al. (2009) related the formation rate of the vortices in chaotic flow regimes with the frequency of oscillation of the jets impingement point.

The chaotic flow regime was also observed in 2D opposed jets geometries studied with CFD simulations by Santos et al. (2005) and Santos et al. (2010), which studied a geometry having a head space and larger chambers in comparison to the jets widths. The headspace is a free space above the injectors where vortices are formed, and these vortices have a key role on the operation at self-sustainable chaotic flow regimes. If the mixing chamber top is directly above the injectors the wall will hinder the flow oscillations and the chaotic flow is not reached, as shown from 2D CFD simulations of Santos et al. (2002). Sultan et al. (2012) and Sultan et al. (2013) made PLIF experiments with deeper mixing chambers than in previous works on T-jets, and considered a head space above the jets and mixing chamber width to jets width larger than six. The deeper chamber, at least 0.5 times the chamber width was also necessary condition for a chaotic flow, on shallow chambers strong wall effect also rendered the flow steady. The flow expansion ratio with the mixing chamber width being at least fourfold the injectors yield was also proven to be a must have condition to onset the vortex street typical of the chaotic flow regime (Sultan et al., 2012). The PLIF experiments corroborated the numerical results of the 2D CFD model. The transition between the two flow regimes in 2D geometries was reported at a Reynolds number of 250. Tu et al. (2014a) and Tu et al. (2014b) provided further experimental evidence

of the existence of a chaotic flow regime with the same vorticity pattern of the 2D CFD model. In this work the 2D CFD model is used at chaotic flow regimes. Further experimental validation is provided in this paper regarding the oscillatory behaviour of opposed jets under chaotic flow regimes.

If the properties of a product depend on the mixing history inside the reactor, which variables can be used to control those properties in a continuous static mixer? The most obvious variable, as proven by Schwarzer et al. (2006) and Kolodziej et al. (1986) is the Reynolds number, and reactor design can also play a key role (Johnson and Prud'homme, 2003 and Sultan et al., 2012). Although, in term of complex products, such as nanoparticles, the final product properties to be targeted go beyond the average particle size, other important properties can be the shape of the PSD or the crystal morphology, which depend in a complex fashion of the mixing history (Silva et al., 2008 and Marchisio et al., 2006). In this paper, additional control of the mixing history and hence on the product properties is targeted using *active mixing*.

Active mixing is the use of a “disturbance generated by an external field for the mixing process” (Nguyen and Wu, 2005), where this disturbance can be of several types, from electrical to mechanical. In this work, the external stimulus is applied to the flow rate of the two feed streams that enter as opposed jets in a mixing chamber, so that the flow rate of each feed stream has a sinusoidal variation over its average value. Active mixing was previously tested on opposed jets mixers, namely: Ito and Komori (2006) on Y-mixers by using a mechanical flow pulsator Komori and Ito (2005); Deshmukh et al. (2000) using a bubble micro-pump; Fujii et al. (2003) using fast switching of the pumps; Zhongliang et al. (2002) using an electrokinetic flow control for Y and T-jets micro-mixers; Li et al. (2008) using a dual syringe pump; and Sun and Sie (2010) using a dynamic pressure signal obtained by varying the level of the feeding liquid in the feeding bottles of two opposed jets. Bierdel and Piesche (2001) studied the effect of the pumps flow pulsation on mixing in an opposed jets mixing chamber of a Reaction Injection Moulding, RIM, machine. Li et al. (2013a) and Li et al. (2013b) studied active mixing in opposed jets modulating the flow rate of air jets with a valve and a speaker, respectively. The previous works on active mixing in opposed jets used tracer experiments to assess the effect of modulation parameters on mixing. In all cases the two fluids entering the mixing chamber from opposed jets flow through parallel paths to the outlet without engulfing each other; the jets modulation set the convective mixing mechanisms that rapidly homogenized both fluids.

In addition to onsetting advective mixing, active mixing can also induce different advection patterns with different mixing scales. Two recent works of Li et al. (2015) and Tu et al. (2015) clearly demonstrate the possibility of controlling the flow and vorticity scales from different flow modulation amplitudes and frequencies. Li et al. (2015) studied the control of flow scales in a CIJ, with a cylindrical chamber, quite similar to those used in RIM. Tu et al. (2015) studied a prismatic T-jets where the flow is quite similar to the one reported from 2D simulations. In both cases the flow oscillations amplitude scale with the external stimuli amplitude and decrease with the

frequency. The 2D simulation results that are presented in this paper will shed some light into the interplay of the external stimuli parameters with the chaotic flow.

In many of the previous works where active mixing was used, the scale of the reactor was in the range of microns to a few millimetres. The control of the mixing scales has been pursued over the last decade through the downscaling of the mixers (Hessel and Löwe, 2003c, Hessel and Löwe, 2003b and Hessel and Löwe, 2003a), active mixing is both an alternative and complimentary route for the control of the mixing scales or to intensify mixing in static mixers. Many micro-reactors operate at low Reynolds numbers, and consequently at steady flow regimes. Steady flow regimes reduce the flow scales for mixing by splitting and recombining flow streams: serial or parallel lamination (Nguyen and Wu, 2005). The reduction of the mixing scales can be faster on flow regimes that exhibit convective mixing mechanisms, such as vortex engulfment (Santos et al., 2005 and Santos and Sultan, 2013). Active mixing is often an option when fast mixing is the goal and it is also a process that can impose the onset of convective mixing in reactors that generally operate at diffusional regimes, see for example the flow regime transition from steady to chaotic in the T-jets mixers studied by Ito and Komori (2006) and in the Y-mixer studied by Deshmukh et al. (2000) when an external stimulus is applied to the feeding flow streams.

From the literature it is clear that mixing and products distribution in opposed jets mixers are strongly affected by operational conditions and the design of the reactor. The challenge of a flexible method to set the mixing scales in a T-jets mixer/reactor is here tackled from the study of the response of the system to an external oscillatory stimulus, in order to obtain different products distributions or properties without redesign of the reactor. In this work, active mixing is promoted over a dynamic flow field at a Reynolds number of 300, so beyond the flow regime transition to a dynamic self-sustainable chaotic flow, but still at a laminar flow regime. The modulation of the jets does not impose the beginning of convective mixing mechanisms, but rather, a new dynamic behaviour on the flow field that is expected to influence the history of mixing between the two flow streams and thus open new prospect for control of the properties of products that are mixing sensitive. Here, the influence on the products properties is not studied; this work is strictly a numerical study focusing on the influence of the jets modulation strategy, modulation frequency and amplitude on the flow dynamics. The effect of the modulation of the jets on the dynamics of the flow is studied by spectral analysis of the dynamic velocity data. The dynamic flow field is obtained from 2D Computational Fluid Dynamics, CFD, simulations using the commercial code FluentTM. This work sets the influence of the active mixing parameters on the flow dynamics and provides the basis for the design of active mixing for opposed jets mixers.

Model

The flow field in a 2D opposed jets mixer was simulated with the commercial CFD code FluentTM using the model of the opposed jets mixing chamber that is shown in Figure 1. This 2D model was previously used for the study of mixing in RIM (Santos et al., 2002, Santos et al., 2005, Li et al., 2007 and Erkoç et al., 2007). The numerical

parameters (e.g., grid density, time interval, residuals, and simulation results) for this 2D model was validated by Teixeira, 2000 and Santos et al., 2010. The set of equations used for the flow field simulation were the Navier-Stokes and continuity equation. The continuity equation for incompressible flow is:

$$\vec{\nabla} \cdot \vec{v} = 0 \quad \text{eq. 1}$$

where $\vec{v}$ is the velocity vector. The Navier-Stokes equation is:

$$\rho \frac{D\vec{v}}{Dt} = -\vec{\nabla} p + \mu \nabla^2 \vec{v} + \rho \vec{g} \quad \text{eq. 2}$$

where p is the pressure, t is the time, $\vec{g}$ the gravity acceleration vector, and ρ and μ are the fluid density and viscosity, respectively. The mass transfer equation

$$\frac{\partial C}{\partial t} + \vec{v} \cdot \vec{\nabla} C = D_m \nabla^2 C \quad \text{eq. 3}$$

was coupled to flow equations, where C is the concentration of a passive tracer and D_m its molecular diffusion coefficient.

Transient state simulations were initiated from an initial solution obtained for steady state conditions. The time interval for the dynamic simulations was set to 10^{-4} s. The numerical grid was made of quadrilateral cells with 0.1mm width. The spatial discretization used was second order upwind for momentum, second order for pressure and the gradients were least-square cell based. The pressure-based segregated solver was set using the PISO algorithm. The time integration scheme was second order implicit.

The 2D chamber is rectangular, having 50 mm length and 10 mm width, connected to 20 mm long injectors, whose centre is located at 5 mm from the close top of the chamber (see Figure 1). The injectors average Reynolds number, Re , is defined for the 2D geometry as

$$Re = \frac{\overline{\rho v_{inj} w}}{\mu} \quad \text{eq. 4}$$

where w is the width of the injectors, which is 1.5 mm, μ is the viscosity, ρ is the density and $\overline{v_{inj}}$ is the time and space average fluid velocity at the injectors. The fluid has $\mu = 20 \text{ mPa} \cdot \text{s}$ and $\rho = 1200 \text{ kg/m}^3$. The no-slip boundary condition was set at the walls, and the outlet was set at constant and uniform atmospheric pressure. The pulsation of the jets is performed according to the following equations:

$$\text{at the left injector} \quad v_{inj}(t)_l = \overline{v_{inj}} + A_l \overline{v_{inj}} \sin(2\pi f t) \quad \text{eq. 5}$$

at the right injector $v_{inj}(t)_r = \overline{v_{inj}} + A_r \overline{v_{inj}} \sin(2\pi f t + \varphi)$ eq. 6

where A_l and A_r are the modulation amplitudes for the left and right jet, respectively, f is the modulation frequency and φ is the phase displacement between right and left injector modulation. The time and space averaged fluid velocity at the injectors is $\overline{v_{inj}} = 3.3333\text{m/s}$.

Eight different cases were simulated and their parameters are shown in Table 1.

- The first case, Case 1, is the Reference Case, where the jets are not modulated, i.e., unforced mixing. When the jets are not modulated the jets impingement point oscillates with a *natural frequency* $\phi = 200\text{Hz}$ (Santos et al., 2009).
- Three different jets' *modulation strategies* were simulated at a fixed amplitude of 0.5 and frequency $f = \phi = 200\text{Hz}$ only one of the jets is modulated (Case2 – single side); the jets' modulation is synchronized, i.e., symmetric, (Case 3 – on-phase); and the jets are modulated out-of-phase (Case 4 – out-of-phase).
- The jets' *modulation amplitude*, $A = A_l = A_r$, was studied in Cases 4 to 6 at a frequency $f = \phi = 200\text{Hz}$ and using an out-of-phase strategy.
- The jets' *modulation frequency*, f , was studied in Cases 4, 7 and 8 at a fixed amplitude of 0.5 and using an out-of-phase strategy.

All the dynamic simulations started from the same segregated steady state solution obtained at $Re = 300$, and were performed thereafter using a fixed time step of 10^{-4}s for a period of 2s. For each time step, the x component of velocity, v_x , was recorded at P1 (jets impingement point: $x = 0$, $z = 5\text{mm}$), P2 ($x = 0$, $z = 22\text{mm}$), and P3 ($x = 0$, $z = 33\text{mm}$), see Figure 1.

The power spectra of the v_x time series were computed using the Discrete Fourier Transform (DFT) that is defined as:

$$G\left(\phi = \frac{k}{N \Delta t}\right) = \sum_{i=1}^{N-1} v_x(i \Delta t) e^{-j2\pi i k / N}, \quad k = 0, 1, 2, \dots, N$$

eq. 7

where $v_x(i \Delta t)$ is the velocity time series, Δt is the interval of the time series, ϕ is the frequency, N is the total number of points in the time series, and $e^{-j2\pi i k / N}$ is the Euler form for a complex number, where j is the imaginary part. The energy in the power spectra of this paper, E , is the modulus of the complex number G .

For the computation of the DFT, the initial period where the flow field in the mixer evolves from the initial steady state solution to a fully developed dynamic state was

not considered. This period was here considered to be 300 ms, more than three times the passage time of the fluids throughout the T-jets mixer.

The evolution of flow through the mixing chamber is assessed from the DFT of the correlation of the time series of v_x at P2 and P3 (cross-correlation). In the cross-correlation the values of v_x at time t_i and point P2, $v_x(P2, t_i)$, are multiplied by the values of v_x at time $t_i + \Delta t$ and point P3, $v_x(P3, t_i + \Delta t)$, and the function is defined as

$$R_{xy}(\Delta t) = \frac{1}{N} \sum_{i=1}^N v_x(P2, t_i) v_x(P3, t_i + \Delta t) \quad \text{eq. 8}$$

The power spectra of the R_{xy} is obtained from the DFT, Equation 6, of the R_{xy} . The typical frequency of the cross-correlation power spectrum, φ , is the inverse of the average delay between the time series at the two locations, and so, considering the distance Δz between P2 and P3, the average velocity of the flow dynamics propagation is given by:

$$\bar{v} = \varphi \Delta z \quad \text{eq. 9}$$

When the flow is an orderly sequence of counter-rotating vortices, the v_x time series recorded at P2 and P3 are two oscillatory functions, having a fixed delay between them. If the vortices are kept exactly the same from P2 to P3, the DFT of the cross-correlation of v_x at P2 and P3 has a well-defined energy peak. The energy peaks can be used to compare the repetitiveness of the flow along the mixing chamber, being the dynamic evolution of the vortices less random when these peaks have higher energy values.

In addition to the spectral analysis of the dynamic data series, which shows the effect of active mixing on the flow dynamics, the energy dissipation will also be computed. The energy dissipation rate is defined as,

$$\varepsilon = 2\nu \langle s_{ij} \cdot s_{ij} \rangle \quad \text{eq. 10}$$

where ν is the kinematic viscosity and s_{ij} is the fluctuating rate of strain tensor. The fluctuating rate of strain is defined as

$$s_{ij} = \frac{1}{2} \left(\frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} \right) \quad \text{eq. 11}$$

where x_i is one of the Cartesian coordinates and $\tilde{u}_i$ is the fluctuating velocity component i and is defined as

$$\tilde{u}_i = v_i - U_i \quad \text{eq. 12}$$

where U_i is the time averaged velocity component i and v_i is the instantaneous velocity component i . The energy dissipation was computed from data obtained from

400 time steps evenly spaced throughout 0.4s of the simulated flow. First, the time-averaged field of the velocity components was computed. After, for each time step, the fluctuating velocity matrix was obtained and the rate of strain tensor computed. The values of ε here reported are the average values obtained from the 400 time steps computed.

The energy dissipation rate is here used due to the relation to the time and spatial scales of mixing, the Kolmogorov time scale that is defined as,

$$\tau_\eta = \sqrt{\frac{\nu}{\varepsilon}} \quad \text{eq. 13}$$

and the Kolmogorov length scale that stands for the smallest hydrodynamic scales in the flow and is defined as,

$$\lambda_\eta = \left(\frac{\nu^3}{\varepsilon}\right)^{1/4} \quad \text{eq. 14}$$

The greater the energy dissipation rate is, the smaller the time and hydrodynamic scales are.

2D model validation

Extensive numerical and experimental validation of the 2D CFD model of the opposed jets mixer used in this study was made by Santos et al., 2010. This model predicts the flow dynamics in opposed jets mixers, as shown by Santos et al., 2009, from the frequency power spectra of the u_x time series obtained both from 2D CFD simulations and Laser Doppler Anemometry (LDA) in a CIJ. The 2D model of Santos et al., 2009 is the same model used in this work, although Santos et al., 2009 used steady boundary conditions at the injectors: Case 1 of this paper. The CIJ mixer used in the LDA experiments is a cylinder with 10mm of diameter and 50mm height having two opposed injectors with 1.5mm diameter. The axis of the opposed injectors is located at 5mm from the top of the CIJ. The 2D model of this work is an axial cut passing through the injectors of the CIJ used in Santos et al., 2009.

Figure 2 shows the power spectrum from LDA data at the impingement point of the jets ($x = 0$, $z = 5.0\text{mm}$). The jets oscillate with typical frequency values in both the 2D model simulation and the 3D CIJ mixer experiments. Furthermore, the oscillation frequency of the jets was related to the same mechanism for both the experiments and the 2D simulations. The jets oscillations frequency was proven by Santos et al. (2009) to have the same value of the formation rate of the vortices downstream the jets.

For a CIJ mixer with the dimensions used by Santos et al., 2009 the imaged vortices have a shape close to an elongated ellipsoid, namely a prolate spheroid, having a polar diameter $d_2 \approx 5\text{mm}$, and an equatorial diameter $d_1 = 3\text{mm}$. Since the ellipsoids are formed in pairs and each vortex only generates half an oscillation, their generated frequency is

$$\varphi_{vortex} = \frac{1}{2} \frac{q_{inj}}{V_{vortex}} \quad \text{eq. 15}$$

where q_{inj} is the inlet flow rate of each jet, and V_{vortex} is the vortex volume

$$V_{vortex} = \frac{\pi d_2 d_1^2}{6} \quad \text{eq. 16}$$

For the experiments that are shown in Figure 2 the Reynolds number was 300, and q_{inj} was $7.3 \times 10^{-6} \text{m}^3/\text{s}$, and so φ_{vortex} is around 150Hz.

For the 2D simulations, as will be later shown in this paper, the oscillation frequency of the jets is around 200Hz. The vortices in the 2D simulations are circular, as can be seen in the streamline maps of Figure 3, having a diameter of approximately 4 mm, using Equation 15 for a 2D geometry,

$$\varphi_{vortex} = \frac{1}{2} \frac{\pi v_{inj} d}{A_{vortex}} \quad \text{eq. 17}$$

where A_{vortex} is the vortex area, for the 4mm vortices $A_{vortex} = 1.3 \times 10^{-5} \text{m}^2$. Recalling that $v_{inj} = 3.3333 \text{m/s}$ and $d = 1.5 \text{mm}$, $\varphi_{vortex-2D} \approx 200 \text{Hz}$.

Although the jets are oscillating with different frequencies in the 2D geometry simulations and in the 3D flow field, the frequency of the oscillations are associated with the same phenomenon: the formation of vortices immediately downstream the jets. The fact that the flow scales determine the frequencies of the jets oscillation was the basis for the present research: if the flow scales set the frequencies, can the flow scales be changed by inducing new frequencies into the system?

Results

The analysis of the results is made for the following parameters of the active mixing: modulation strategy; amplitude of the modulation; and frequency of the modulation. For each case the following results are shown: the streamline maps at a time instant with the plots of $v_x^* = v_x / \overline{v_{inj}}$ time histories at P1 in Figure 3 for cases 1 to 4 and Figure 4 for cases 5 to 8; the power spectra of v_x time histories at P1, P2 and P3 in Figures 5, 6 and 7 respectively; and the power spectra of the cross-correlation of v_x time series between P2 and P3 in Figure 8. Figure 9 shows the maps of energy dissipation for all the cases. The maximum energy of the power spectra of v_x time series at P1, P2 and P3 and from its cross-correlation between P2 and P3 are listed in Table 2. Also in Table 2 are listed the values of energy dissipation rate for each case.

Reference Case

The main features of the undisturbed flow field at $Re = 300$, Case 1, are described first in order to set the Reference Case that will be used to assess the effects of the different active mixing parameters. The overall flow patterns of the unforced flow field are seen from the streamline maps of Figure 3. Two opposite jets impinge at the mixing chamber axis and after this point a vortex street is formed. The jets slightly bend towards the outlet in their path from the injector to the impingement point; as a consequence the jets' impingement point actually occurs around $z = 7.5 \text{mm}$, i.e.,

slightly below the injectors' and chamber's axes intersection – P1 ($x = 0$, $z = 5\text{mm}$). After impinging on each other, both jets follow a parallel path, keeping their initial velocity while flowing around the vortices throughout the chamber until the outlet; this is seen from the streamlines that keep roughly equidistant from the injectors to the outlet. The dynamic evolution of these vortices down the mixing chamber promotes an intense and fast mixing of the two fluids (Santos et al., 2005).

The mixing chamber has a closed cavity at the top of the jets where two round vortices are formed, see Figure 3. Santos et al. (2002) showed that these vortices have a key role on the sustainability of a dynamic flow field inside a T-jets mixer. When the upper cavity is removed, the flow field can only have some dynamic evolution if an external perturbation is applied to the jets, but when the perturbation ceases the flow quickly tends to one of the five steady states identified by Santos et al. (2010). This cavity is needed to operate the opposed jets mixer at chaotic flow regime.

The impingement point of the jets oscillates, as previously reported by Wood et al. (1991), Johnson and Wood (2000), Teixeira et al. (2005), Erkoç et al. (2007), and Santos et al. (2008), which causes the time histories of v_x at P1 to oscillate as shown in Figure 3. The frequency of the jets impingement point oscillation is around 200 Hz, as can be seen from the energy peaks of the power spectra of v_x at P1 in Figure 3. These oscillations are self-sustainable, i.e. they are not kept by any external perturbation but feed themselves from the instabilities generated when two well balanced high speed jets are stopped against each other in a short distance of 5 mm. The $\phi = 200\text{Hz}$ value is here called the natural oscillation frequency; the modulation frequencies are set equal to $f = \phi$ in most cases. The two exceptions are Case 7 and Case 8 where the modulation frequencies values are $f = 2\phi$ and $f = \phi/2$, respectively.

The energy peak $\phi = 200\text{Hz}$ is caused from the formation of the two vortices located immediately downstream the jets, each vortex has a diameter less than half the mixing chamber width. These vortices formation rate is equal to $\phi_{\text{vortex-2D}} \approx 200\text{Hz}$ Santos et al. (2009).

For Case 1, Figure 5 shows that the power spectrum presents an overall spread of energy around the 200Hz frequency. The jets impingement point is not oscillating with a single frequency but within a range of frequencies, a clear signature that this flow field has a chaotic behaviour. From the dynamic characteristics of the described flow field at $Re = 300$ this flow regime was classified as *self-sustainable chaotic flow regime* in previous works of the present research team (e.g. Santos et al., 2005, Santos et al., 2008 and Erkoç et al., 2007).

The small vortices initially formed downstream the jets evolve to vortices that occupy the whole width of the mixing chamber. These fully developed vortices flow in an orderly sequence, forming a vortex street. At P2 and P3 the energy peaks are around 72 Hz. One wavelength of the oscillation in the v_x time history requires the passage of a pair of counter rotating vortices (Santos et al., 2009). The energy spread around the 72 Hz peak is directly associated to the repetitiveness of the vortices evolution in the

mixing chamber. If all the vortices are equal and pass through P2 and P3 exactly the same way, then the power spectra will present very well defined peaks of energy. On the other hand, if the vortices have more random distributions of size and shape then the resulting power spectra will exhibit an energy spread around a typical value. Complete details of this analysis are given in Santos et al. (2009).

The repetitiveness of the vortices passage between two separated points can be assessed from the cross-correlation of the u_x time histories at those points. If the period for the vortices to travel from P2 to P3 is almost constant, i.e., a very well ordered flow throughout time, the power spectrum of the cross-correlation of u_x has a well-defined energy peak, while more random or chaotic evolutions render a broader spread of energy around the typical frequency. Furthermore, the cross-correlation is a series that oscillates with a period directly related to the time a vortex takes to go from P2 to P3 (Equation 8). Here the value $\phi = 144\text{Hz}$ corresponds to a propagation velocity throughout the mixing chamber axis of 1.6 m/s, i.e., half the average velocity of the jets, $\overline{u_{inj}} = 3.3333\text{m/s}$, and higher than the average velocity in the mixing chamber that is approximately 1 m/s.

For the Reference Case the energy dissipation rate reaches the highest values around the point where the jets impinge. The ϵ values decrease from the impingement point towards the outlet although keeping local maxima in the chamber axis. Upstream the jets' impingement point and also in the region crossed by the opposed jets the flow has only small oscillations and so the turbulence intensity values are small. Liu and Fox (2006), Santos *et al.* (2008) and Lince et al. (2011) had previously reported that the turbulence values peak at the impingement point in a confined impinging jets mixer, and reported similar overall distributions of turbulence related quantities.

Modulation Strategy

Four different modulation strategies were defined: Case 1 – no modulation; Case 2 – only one of the jets is modulated; Case 3 – the jets are modulated with a synchronised flow rate oscillation; and Case 4 – the jets are modulated with out-of-phase flow rate oscillation. The energies of the typical frequencies of the flow in the mixing chamber for these cases are shown in Figures 5 to 8 and listed in Table 2.

The lowest energy values at the typical frequencies are observed for the on-phase pulsation case – Case 3. Most of the energy values are smaller than those in the undisturbed case, except the energy peak of the cross-correlation. For Case 3, since both jets are modulated simultaneously, their strengths are kept balanced, so there is no disturbance of the jets equilibrium. When a jet loses its strength, it is not compensated by the opposed jet, which is also retracted, conversely when a jet increases its strength it is matched by the opposed jet.

For Case 3 the energy level associated with the jets oscillations at the impingement point, P1, presents the lowest energy of all studied cases (Table 2) and it is five times lower than the Reference Case. Although the jets are modulated at the natural frequency value, $f = \phi$, this is done symmetrically and thus, instead of enhancing the

flow dynamics, this modulation strategy is opposing the flow dynamics by favouring the balancing of the opposite forces: the jets.

For Case 3 it is also observed from the streamline maps in Figure 3 that there is not a clear vortex street below the impingement point of the jets, and so the energy peaks associated with the flow field dynamics at P2 and P3 are dimmed. The jets oscillations and the formation of the vortex street are intimately associated mechanisms (Santos et al., 2009), and so the decrease on the dynamics of one mechanism also decreases the other. This result is further evidence of the relation between vortex formation rate and jets oscillation frequency.

Modulating only one of the jets, Case 2, favours the flow dynamics only at the jets' impingement point, P1, where the energy peaks are higher than for the Reference Case 1 (Table 2). At the jets impingement point another frequency peak appears between 50 and 100Hz (Figure 5), which is at the same frequency values associated with the formation of the fully developed vortices, at P2 and P3. Nevertheless, the energy levels in the range of 50 to 100Hz at P2 and P3 are approximately kept in Case 2, when compared to the undisturbed flow dynamics (Figures 6 and 7). The vortex street repetitiveness is thus not enhanced with the modulation of a single jet. In addition, at P2 (Figure 6) the highest energy peak is identified at 200 Hz, i.e., at the same frequency of the jets natural oscillations and of the jets modulation. This energy peak almost vanishes at P3 (Figure 7). The jets single side modulation is thus enhancing the oscillations of jets at the impingement point with the associated formation of the vortices that occupy half the mixing chamber. For Case 2, these vortices are the predominant flow structure until further downstream than for the Reference Case.

The modulation of the jets out-of-phase clearly enhances the flow dynamics repetitiveness with the increase of all energy peaks identified from the Reference Case. The out-of-phase modulation of the jets imposes itself to the flow own dynamics most of the time, as can be seen from the time histories in Figure 3, where there is a prevalence of the periods of higher amplitude oscillations that are induced from the imposed modulation. This is the only modulation strategy where a clear prevalence of the imposed dynamics over the natural dynamics is observed from the v_x time histories at P1.

The enhanced regularity of the vortices evolution can also be observed from the increase of the energy peaks at P2 and P3 (Table 2 and Figures 6 and 7). For Case 4, although the modulation dynamics imposes itself clearly to the flow natural dynamics, since the values of the modulation are those of the natural frequencies, $f = \phi$, in the end this is translated by the enhancement of the flow own dynamics.

The frequencies of the cross-correlation are identified in Figure 8 and Table 2 (P1×P2). The frequency values decrease only slightly for the cases where the jets are modulated, and so the propagation velocities are kept within the same values. The major difference is in the energy values for Case 4 that are one order of magnitude higher than in the other strategies. The strategy of Case 4, out-of-phase modulation,

gives further order to the system yielding the most repetitive vortex street of the four analysed strategies.

The modulation strategies have distinct impact over the flow dynamics, with some cases imposing to the flow own dynamics much more clearly than others do. The cases that can impose more clearly are those that more markedly break the symmetry of the opposite balanced forces, the best example being Case 4, out-of-phase modulation. The jets modulation strategy promotes flow regimes that range from chaotic behaviour that this flow naturally presents, to a more orderly oscillatory behaviour that is imposed to the system from the sinusoidal stimulus.

Modulation Amplitude

For the strategy that best imposes to the flow natural dynamics, out of phase, three amplitudes were tested, $A=0.25$, 0.5 and 1 , corresponding to Case 5, Case 4 and Case 6, respectively. These cases are compared to the Reference Case, Case 1, where $A = 0$.

For Case 5, where the modulation amplitude is lower, $A = 0.25$, the flow own dynamics is dimmed from the imposed modulation. Except for the jets impingement point oscillations, P1, all other energy levels are lower than in the Reference Case. The jets oscillations are slightly favoured in Case 5, although the modulation does not impose to the natural oscillations so clearly as in Case 4, as can be seen from the time histories of v_x in Figures 3 and 4. Nevertheless, the associated energy peak is higher than in the Reference Case (Figure 5). In both cases 4 and 5, the jets oscillations frequencies are prevailing up to P2, and thus both cases favour the formation of the smaller vortices occupying half the mixing chamber. The energy peaks at the typical frequencies for Case 5 at P2 and P3 are slightly lower than in the Reference Case (Table 2 and Figures 6 and 7). For the power spectra at P3 and P1×P2, which are related to the orderly evolution of the vortices throughout the mixing chamber, the energy peaks are kept within the same frequencies but at lower values. Thus in the cases with lower oscillation amplitudes the imposed oscillation interact with the flow natural/unforced dynamics but without overriding it, and so in some locations the flow dynamics is enhanced, P1, but on others it is dimmed, e.g., P3.

As the amplitude is increased, the imposed dynamics becomes dominant and the energy peaks at the power spectra have higher values and less energy spread. The flow becomes increasingly ordered with the imposition of the oscillatory movements of the jets impingement point over the natural chaotic oscillations. When the amplitude is $A = 1$ the flow is completely dominated from the modulation of the jets and the vortex street evolves quite orderly as can be seen from the streamline maps of Figure 4, consequently the power spectra at P2 and P3 present higher energy peaks.

For Case 6, where $f = \phi = 200\text{Hz}$ and $A = 1$, a frequency phenomenon around 60 Hz , is distinctly identified at the impingement point of the jets. Although the oscillations of the jets are being forced with an amplitude equal to the average flow rate, $A = 1$, the frequency phenomenon associated to the fully developed vortices

passage still causes a measurable influence at the jets impingement point. This energy peak is also clear in the Case 4, $A=0.5$, and gives further evidence that the jets oscillations are being driven from the vortices formed downstream, which was postulated as the main mechanism for the jets impingement point oscillation in Santos et al., 2009.

The modulation amplitude sets the relative weight of the imposed dynamics over the flow natural dynamics. With lower amplitudes there is an interaction between the imposed and the natural dynamics: some dynamic behaviours are enhanced while others are dimmed. When the amplitude of the modulation increases, the dynamics of the flow becomes increasingly defined from the imposed behaviour, and so the flow dynamics presents a trend going from a chaotic behaviour to a purely oscillatory dynamics.

Modulation Frequency

Three different modulation frequencies were studied using the out-of-phase strategy and $A=0.5$: the jets were modulated with the same frequency of the jets impingement point natural oscillations, Case 4 $f = \phi = 200\text{Hz}$, at double this value, Case 7 $f = 2\phi$, and at half this value, Case 8 $f = \phi/2$.

When the modulation frequency is doubled, Case 7, it becomes too fast to impose an effect over the flow own dynamics and the energy levels in the power spectra at P1, P2 and P3 are the lowest among all cases. The high frequency modulation does not enhance any of the natural flow behaviours and so it mainly opposes to the natural dynamics of the flow. Okkels and Tabeling (2004) obtained similar results in their work on the quantification of folding, or area generation, between two initially segregated fluids flowing through a channel with two opposite lateral ports. From the lateral ports a flow oscillation was imposed, and folding was observed to increase with the oscillation amplitude but decrease with the oscillation frequency. Okkels and Tabeling (2004) stated that inefficient mixing occurred when “the perturbation of a point on the interface during the first half of the mixing region is completely reversed during the last part, resulting in vanishing net perturbation.” Zhongliang et al., 2002 that modulated the flow in Y and T-jets mixers to form bands of two saline buffers also observed that the modulation effect decreases with an increase of its frequency. At low frequency, consecutive bands of the different buffers are formed. However when the modulation frequency is increased, the flow tends to the case of two segregated streams flowing in parallel. Qiu et al. (2012) and Li et al. (2013b) also observed using 2D CFD simulations of opposed jets that mixing improves with the amplitude of the external stimulus and for higher frequency values the effect of the pulsation is dimmed. In the present case the imposed perturbation, e.g., an increase of flow rate in the right jet, is reversed too fast from the opposite jet to cause an effect on the flow dynamics.

The modulation using $f = \phi/2$, Case 8, causes an earlier formation of the fully developed vortices that onset at P1. In Case 8 at the jets impingement point the flow oscillates with the modulated frequency, $f = \phi/2$, and the natural oscillation

frequency, $\phi = 200\text{Hz}$, is no longer identifiable. The imposed frequency is also dominant at P2 and P3, where the frequency associated to fully developed vortices passage is no longer around 60 Hz as on the other cases but it is now 100 Hz. Thus, the scale of the fully developed vortices is tuned when the modulation frequency is close to the values of the fully developed vortices natural oscillation frequencies. The passage rate of the vortices is given by Santos et al., 2009

$$\varphi_{\text{vortex-2D}} = \frac{4\overline{v_{inj}}w}{\pi D_{\text{vortex}}^2} \Leftrightarrow D_{\text{vortex}} = \sqrt{\frac{4\overline{v_{inj}}w}{\pi\varphi_{\text{vortex-2D}}}} \quad \text{eq. 17}$$

where D_{vortex} is the diameter of the vortices.

A 60 Hz frequency would correspond to vortices completely occupying the chamber width, i.e., $D \approx 10\text{mm}$, while at 100 Hz the vortices' diameter is around $D \approx 8\text{mm}$. The propagation velocity between P2 and P3 at Case 8 is 2.1 m/s, from Equation 4 and considering the frequency of the cross correlation: $\varphi = 194\text{Hz}$. For Case 8, all the energy peaks associated with the vortex street present higher energy values, and the flow is more repetitive. Modulating the jets at 100 Hz yielded a very repetitive flow with a lower scale for the vortices that evolve at higher velocities throughout the mixing chamber.

The control of vortex scales in a vortex street from the modulation of a stream was also observed by Wang (2003) and Wang (2006) using an apparatus that joined two parallel streams of water being one of them oscillated using a loud speaker according to a patented apparatus Fiedler and Wang (2002). In a widely used mixing geometry, such as the T-jets mixer, this is the first numerical work reporting a control of flow / mixing scales by means of the parameters of active mixing. Previous works have mainly focused on promoting the transition from segregated flow regimes to regimes having convective patterns that engulf both flow streams (Zhongliang et al., 2002, Ito and Komori, 2006, Sun and Sie, 2010, Qiu et al., 2012, Li et al., 2013a, Li et al., 2013b), these works focused on the effect of active mixing on mixing of tracers and not on the dynamics of the flow.

Recently two works, one on CIJs and the other on T-jets focused on the control of flow regime and mixing scales from the jets pulsation (Tu et al., 2015, Li et al., 2015). On both cases was clear a larger influence of the external stimuli for lower excitation frequencies. The flow patterns obtained by Tu et al. (2015) are quite similar to the 2D CFD model results in this paper. The 2D simulation results provide a framework for the explanation of Tu et al. (2015) results. Tu et al. (2015) studied three T-jets configurations, and configuration II has particular geometrical relations very close to the 2D model here used: $D/w = 6$, $h/d = 5$. Furthermore the chamber depth was $4D/3$, which ensures that there is no wall effect and thus chaotic flow regimes were reported at Reynolds number 210. When the excitation frequency is considerably lower than the natural oscillation frequency, the plots show the simultaneous existence of both frequencies, with a low frequency large amplitude movement of the jets impingement point position overlapped with the higher frequency oscillations of the flow. This was remarkably clear for the chaotic flow regime at $\text{Re}=210$, where Tu et al. (2015) results show a natural frequency of 20Hz and when excited at 1Hz the addition of both oscillation frequencies was clear. For excitation frequencies closer to

20Hz, for example 10Hz and 25Hz from Tu et al. (2015) results is clear a single oscillatory behaviour with a frequency set from the external stimulus.

Steady flow regimes for $Re=70$ were also studied by Tu et al. (2015), and the onset of vortices having a diameter close to the mixing chamber diameter was clear at an excitation frequency of 5Hz. This 5Hz frequency corresponds to the Strouhal number of the natural oscillation frequency for chaotic flow regimes. Larger excitation frequencies, 25Hz, over steady states give rise to smaller vorticity scales that are not able to engulf fluid from both injectors originating a segregation plane in the middle of the mixing chamber.

Mixing assessment

In the present work, mixing is assessed from the values of the energy dissipation rate, ε . The contour plots of ε , which were computed from the 2D CFD simulations in the simulation period 0.4 s to 0.8 s using a total of 400 time steps evenly sampled, are shown in Figure 9. Table 2 has the spatial average values of the energy dissipation rate, $\bar{\varepsilon}$.

The space averaged energy dissipation, $\bar{\varepsilon}$, rises at least 30% when an external stimulus is applied to the system. Although the distribution patterns of ε depend on the pulsation strategy applied to the system. In Case 1, ε has higher value in the region where the jets impinge on each other decaying outwards of that region. Although the distribution of the energy dissipation is not as concentric as in CIJs (Liu and Fox, 2006) the same overall trend of a clear concentration of the larger values of ε around the impingement point is observed in both T-jets and CIJ geometries.

The cases where pulsation is imposed to the system show an increase of $\bar{\varepsilon}$, however the maps of ε show that this increase is not concentrated in the jets impingement region. In most cases ε is increased mainly in the jets path immediately after the inlet. When the flow is not modulated, the jets are steady until approximately the region where they impinge. The modulated jets velocities have an imposed oscillatory component, and so this results in an increase of $\bar{\varepsilon}$. The flow oscillation is only contributing to mixing after the jets impinging point, and so the increase of ε in the jets prior to the impingement point do not stand for an actual increase on mixing. Because the amplitude of the modulation of the inlet flow rate has direct impact on the value of $\bar{\varepsilon}$, only the cases with equal amplitude of pulsation are directly comparable from this quantity. The flow rate modulation strategy is also important; while in out-of-phase modulation the net variation of flow rate at the chamber outlet is zero, for both the on-phase pulsation and single phase-pulsation there is an actual oscillation of the flow rate throughout the mixing chamber. Although, the only strategy where the net oscillation of flow rate is zero is the out-of-phase strategy, the values of $\bar{\varepsilon}$ are close to those of the single-phase and on-phase oscillation.

For the modulation frequency, considering the same amplitude, the values of $\bar{\varepsilon}$ do not show significant differences, although the spatial distribution of ε is different for the three frequencies: $\phi/2$, ϕ and 2ϕ . The case where the system was observed to engage

into resonance with the modulation frequency, $\phi/2$, has higher values of ε around the impingement point than in the ϕ and 2ϕ cases.

The amplitude only has a clear impact on the $\bar{\varepsilon}$ value for higher values, $A = 1$, where the system dynamics is mainly ruled from the modulation of the inlet flow rate.

Further insight into mixing was gathered from mass transfer simulations coupled to the flow field. Figure 10 shows contour plots of the concentration of a passive tracer injected through one of the opposed jets. At the outlet there is no clear difference of tracer homogeneity between the different cases. In these simulations the Schmidt number is around 5.7, and is defined as

$$Sc = \frac{\mu}{\rho D_m} \quad \text{eq. 18}$$

Typical Schmidt numbers for liquids are at least two orders of magnitude larger than the value used in this work. Thus, these results are affected by a large diffusion value, which makes the tracer concentration unrealistically homogeneous. Diffusion free simulations using VOF model that were introduced by Fonte et al. (2010) would have to be made to assess the impact of modulation in the mixing scales.

Nevertheless, from the concentration maps is possible to identify the mixing mechanisms. Furthermore, from the knowledge of the influence of the jets flow rate modulation on flow dynamics it is possible to identify its effect on mixing. A clear image of the effect of modulation on mixing is only possible from the dynamic visualization of the tracer maps, which is not presented in this paper, although movies of these simulations were made and used by the authors on this analysis.

The vortices formed immediately downstream the jets merge into larger vortices: fully developed vortices that engulf fluid from both jets. From Figure 10 is clear that fluid entering the chamber is initially engulfed into the smaller vortices, immediately downstream the jets. After these smaller vortices having a diameter around half of the mixing chamber width, detach and evolve into fully developed vortices that occupy the complete width of the mixing chamber. Santos et al., 2005 proved that the fully developed vortices are the main mixing mechanism in these mixers; the earlier these vortices are formed the earlier both fluids will mix. From spectral analysis, it is clear that the two cases where fully developed vortices dominate through larger areas of the mixing chamber are Case 6, modulation amplitude equal to one, and Case 8, modulation frequency $\phi/2 = 100\text{Hz}$. These are the two cases where mixing is most likely occurring earlier.

In the movies from Case 6 is clear the earlier formation of fully developed vortices closer to the jets impingement point, which is also seen in Figure 10 as well as the earlier homogenization of fluid. The vortices centres are alternately rich in fluid from left and right injector; this is visible in maps from different cases in Figure 10. For Case 6 the vortices centre local maximum concentration disappears earlier in the mixing chamber.

Case 8 has similar dynamics to Case 6, although the effect of jets modulation on concentration fields is not as dramatic as in Case 6, the energy used in Case 8 is smaller than in Case 6. Jets flow rate modulation in Case 8 is making the system to go into resonance, i.e. in Case 8 the excitation frequency causes the system to oscillate at its natural frequencies but with higher amplitude, this is accomplished in mechanical systems by setting an excitation frequency to values close to the system's natural frequencies.

Conclusions

The control of mixing was studied in an opposed jets mixer by means of the modulation of the jets flow rate. The study focused on the impact of the jets flow rate modulation on the vortex formation and flow field dynamics.

The pulsation strategy has a strong impact on the flow dynamics. While out-of-phase strategies clearly affect the flow own dynamics, the other strategies tend to interact more with the flow own dynamics or even to oppose it.

The amplitude of the modulation can be used to set the weight of the imposed dynamics over the natural dynamics. Low amplitudes only affect the flow dynamics while higher amplitudes can completely determine it.

The frequency of pulsation is shown to have a great impact on the flow dynamics. With values close to the natural jets oscillation frequencies the jets modulation enhances the regularity of the system resulting in a clear oscillatory behaviour of the jets at the impingement point. Imposed oscillation values, close to the natural frequency generated by passage of the fully developed vortices, can force the frequency of the fully developed vortices passage and make it more orderly. The energy of the flow oscillations is increased from an excitation given by an external oscillatory stimulus that has a frequency close to the flow natural frequencies – resonance. This study clearly shows the importance of knowing the flow natural frequencies when setting the frequency of an external stimulus and the possibility to induce a flow field in opposed jets mixers into a resonant state. Eq. 17 of this paper enables a prediction of the flow natural frequencies that can be used on the design of active mixing systems for opposed jets mixers. The conclusions of this paper are likely to be applicable to other mixers operating under transition flow regimes where the energy containing flow scales are set by the reactor geometry.

Acknowledgements

This work was co-financed by FCT and FEDER under Programme PT2020 (Project UID/EQU/50020/2013) and by QREN, ON2, FCT and FEDER (Projects NORTE-07-0162-FEDER-000050 and NORTE-07-0124-FEDER-000013). Ertugrul Erkoç and Cláudio Fonte acknowledge their Ph.D. scholarships by FCT, with references SFRH/BD/18901/2004, and SFRH/BD/39038/2007, respectively.

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Figure 1 2D model used for the CFD simulations of the active mixing on T-jets mixers.

Figure 2 Power spectra of u_x obtained from LDA data at $Re = 300$.

Figure 3 Streamlines of flow field in the mixing chamber from the dynamic simulations at average $\text{Re}=300$, using different pulsation strategies, and time histories of v_x^* at $\text{Re}=300$ at P1, for Case 1 to 4.

Figure 4 Streamlines of flow field in the mixing chamber from the dynamic simulations at average $Re=300$, using different pulsation strategies, and time histories of v_x^* at $Re=300$ at P1, for Case 5 to 8.

Figure 5 Power spectra from time histories of v_x^* for Case 1 to 8 at $Re=300$ at P1.

Figure 6 Power spectra from time histories of ν_x^* for Case 1 to 8 at $Re=300$ at P2.

Figure 7 Power spectra from time histories of v_x^* for Case 1 to 8 at $\text{Re}=300$ at P3.

Figure 8 Power spectra from the cross-correlation of the time histories of v_x^* between P2 and P3, for Case 1 to 8 at $Re=300$.

Figure 9 Maps of energy dissipation obtained from the 2D CFD simulations in the interval $t = 0.4$ to 0.8 s Case 1 to 8 at $Re=300$.

Figure 10 Maps of species concentration from the 2D CFD simulations coupled with mass transfer for Case 1 to 8 at $Re=300$.

Table 1 Summary of simulated cases

Case Number	Modulation Strategy	Amplitude and Frequency
1	No modulation	$A = 0$
2	Single side modulation	$A_l = 0.50, f = \phi$
3	On-Phase	$A_l = A_r = 0.50, f = \phi, \varphi = 0$
4	Out-of-Phase	$A_l = A_r = 0.25, f = \phi, \varphi = \pi$
5	Out-of-Phase	$A_l = A_r = 0.50, f = \phi, \varphi = \pi$
6	Out-of-Phase	$A_l = A_r = 1.00, f = \phi, \varphi = \pi$
7	Out-of-Phase	$A_l = A_r = 0.50, f = 2\phi, \varphi = \pi$
8	Out-of-Phase	$A_l = A_r = 0.50, f = \phi/2, \varphi = \pi$

Table 2 Maximum energy values from the power spectra of the u_x time histories at P1, P2, P3, and from the cross-correlation of the u_x time histories at P2 and P3 (Figures 5 to 8), and the rate of energy dissipation.

Case Number	Maximum energy				$\bar{\varepsilon}$ $\times 10^{-1}$ W/kg
	P1 $\times 10^3$ m	P2 $\times 10$ m	P3 $\times 10$ m	P2 $\times$ P3 $\times 10^{-6}$ m ² /s	
1 – no modulation	8.4	2.9	3.5	1.5	13
2 – one side modulation	25	2.5	4.5	2.5	17
3 – on-phase	1.7	1.3	2.5	2.0	18
4 – out-of-phase $A=1/2, f=\phi$	57	4.1	7.2	19	17
5 – out-of-phase $A=1/4, f=\phi$	53	1.9	2.1	1.4	20
6 – out-of-phase $A=1, f=\phi$	58	9.8	9.4	48	34
7 – out-of-phase $A=1/2, f=2\phi$	9.2	2.0	2.4	36	21
8 – out-of-phase $A=1/2, f=\phi/2$	3.7	2.0	6.5	65	24