

Mixing Studies with Impinging Jets

PIV/PLIF Experiments and CFD Simulation

a Ph.D. Dissertation in Chemical and Biological Engineering by
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Summary

The present thesis concerns the identification of the mixing mechanisms and flow regimes in a Confined Impinging Jets (CIJs) mixer with typical dimensions of a mixing chamber of a Reaction Injection Molding (RIM) machine. The lamellar microstructure of the flow arising from mixing of the two impinging jets is characterized. The jets' interaction under balanced and unbalanced flow conditions is also studied.

Mixing length scales and the stretching properties of the impinging jets flow are studied from 3D Computational Fluid Dynamics (CFD) simulations coupled with a Lagrangian tracking of the position and deformation of passive material fluid elements. The lamellar structure of the flow is also studied from an Eulerian-Eulerian approach with a 2D non-diffusive two-phase CFD model of the flow coupled with a dynamic grid adaption algorithm. Mixing patterns in the flow are obtained and studied experimentally with the Planar Laser Induced Fluorescence (PLIF) technique. These studies allow the clear identification of the mixing mechanisms in the impingement jets flow. Operational parameters such as the jets Reynolds number and kinetic energy rate ratio are. An elastic analogy of the two impinging jets behavior is used to predict the impingement point position under different flow conditions, showing good agreement with CFD simulations of the flow and results from Particle Image Velocimetry (PIV) experiments.

Experimental and numerical results show that advective dynamic mixing in the impingement jets flow occurs above a critical Reynolds number equal to 110. For optimal mixing efficiency, the impingement mixing must occur at the center of the mixing chamber, setting the kinetic energy rate ratio between the two jets as estimated by the proposed elastic analogue model, and for $Re > 110$. The injectors' diameter of the mixer can be adapted to comply simultaneously with reaction stoichiometry. The present dissertation offers deeper insight on how impingement mixing in RIM produces striation length scales sufficiently thin for the rate of polymerization reaction in the mold to become chemically controlled. It characterizes as well the system behavior under unbalanced jets flow conditions in order to offer robustness to polymer production in RIM from monomers or pre-polymers formulations with different physical properties or stoichiometric ratios different from one.

Introduction and State of the Art

Over the last decades, mixing by opposed impinging jets appeared as an alternative to conventional mixing in stirred tanks and has revealed to be well suited for continuous processes that require a rapid homogenization of streams (Siddiqui et al., 2009). In this technology, two or more reactants are injected into a confined space, the mixing chamber, through opposed jets. The rapid deceleration and quite strong energy dissipation of the two jets, after impingement in the confined space, allows for mixing to be achieved rapidly and without movable mechanical parts, such as stirring devices. Opposed impinging jets mixers/reactors are generally divided into two types, depending on its geometry: Confined impinging jets mixers with cylindrical chamber and injectors (Mahajan and Kirwan, 1996; Johnson and Prud'homme, 2003; Sohrabi and Zareikar, 2005; Marchisio, 2009; Siddiqui et al., 2009; Icardi et al., 2011) and T-jets mixers with rectangular cross-section chamber and injectors (Engler et al., 2004; Bothe et al., 2006; Gradl et al., 2006; Bothe et al., 2008; Adeosun and Lawal, 2009; Gradl and Peukert, 2009). These devices have a wide range of applications in different industrial processes such as precipitators for nanoparticle production or in reactive polymer injection (RIM).

Although the results of the present study are relevant for the different applications of impinging jets mixing, Reaction Injection Molding technology is the focus of this dissertation as the case study for the application of this mixing method. This is due to its economic relevance and growing interest in applications for the automotive and aeronautical industries. The need for solutions that turn the process more robust is the major driving force for this work.

Reaction Injection Molding is an industrial process for the production of plastic parts conceived in the laboratories of Bayer AG in Leverkusen in 1964 (Berins, 1991). In this process, two or more viscous liquid monomers or pre-polymers are injected and mixed in a cylindrical mixing chamber by opposed impinging jets. The mixture that leaves the mixing chamber is discharged into a mold where the polymerization occurs and the plastic part is formed. Efficient mixing of the two monomers, during the residence time inside the mixing chamber before entering the mold, is crucial in order to avoid zones of unreacted monomers that may lead to structural defects and the eventual rejection of the plastic parts (Kolodziej et al., 1986). The injected fluids have viscosities in the range of 10 to 1000 mPa.s and are introduced in the cylindrical mixing chamber through injectors with 1–3 mm of diameter at high pressure (100 to 200 bar) and velocities in the range of 10 to 150 m/s. The mixing chamber is typically a cylinder with 3-15 mm diameter and length around 5 times the diameter (Oertel, 1985). The mean residence time in the mixing chamber is in the range of 10 – 100 ms.

Mixing from impinging jets occurs due to the stretching and folding of the two streams in the mixing chamber promoted by a hydrodynamic instability. The result is the formation of a lamellar structure out of the fed streams. Figure 1 shows an image of the lamellar structure of the flow resulting from impingement mixing obtained from Planar Laser Induced Fluorescence experiments. In the particular case of RIM, due to the short residence times (10 to 100 ms) inside the mixing chamber, molecular diffusion is not expected to occur significantly. Instead, distances between the two initially segregated monomers or pre-polymers must be reduced rapidly in the mixing chamber by advection mechanisms. Later in the mold, diffusion must homogenize the mixture at a molecular level in a time scale smaller than the reaction time scale. For the rate of polymerization reaction to become chemically controlled, the formed lamellar structure must have striation thicknesses, s , of the order of 10 μm (Lee et al., 1980). The striation thickness in a binary layered mixture of components A and B is defined as

$$s = \frac{\delta_A + \delta_B}{2}, \quad (1)$$

that is, half the sum of the local laminae thickness of each component, δ_A and δ_B .

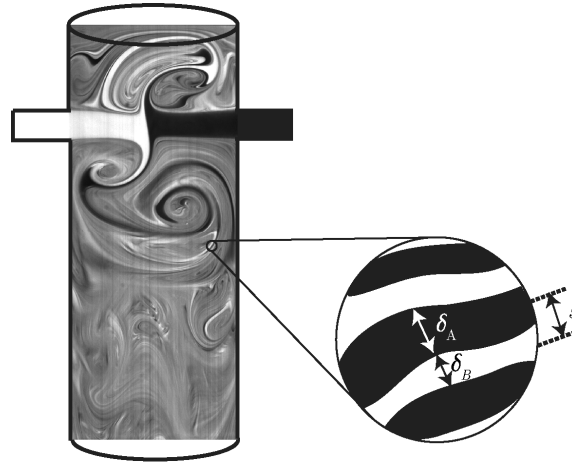


Figure 1. PLIF image of impingement mixing of a binary mixture.

Lee et al. (1980) proposed a model to estimate an average striation thickness, \bar{s} , in RIM's lamellar structure, considering that the impingement process is, on a local scale, a rapid, two dimensional stagnation flow, and assuming that the stretching remains in the plane of each lamina. Tucker III and Suh (1980) proposed, using isotropic turbulence theory considerations, that an average striation thickness in RIM is determined instead by the smallest hydrodynamic length scale of the flow, the Kolmogorov scale. Tucker III and Suh (1980) state that mixture clumps smaller than smallest hydrodynamic length scale are completely contained in a single eddy motion, thus not experiencing shear deformation. The models of Lee et al. (1980) and Tucker III and Suh (1980) for the mean striation thickness show some similarities, such as a dependency on the geometrical mixing chamber parameters and the formation of smaller lamellae thicknesses as Re is increased. However, each model predicts substantially different values for the striation thickness. The two models main limitation resides in the fact that flow heterogeneities and different residence times inside the mixing chamber will produce a distribution of striation scales, which both fail to provide. In fact, the quality of the final plastic part will be mostly influenced by the largest lamellae. Large striation structures may not react in the required time due to diffusional limitations and lead to structural or aesthetical defects. Studies on the effect of a distribution of striation thicknesses in the polymerization reaction can be found in Fields and Ottino (1987a) and (1987b), and Sokolov and Blumen (1991a) and (1991b).

Kolodziej et al. (1982) measured experimentally a distribution of striation thickness in the actual formed plastic and its dependence on the Reynolds number by mixing and letting react a polyurethane formulation in a RIM machine with typical dimensions, marking the polyol reactant with carbon black. With microscope visualizations of the lamellar structure in the formed plastic, a striation thickness histogram was determined for each Reynolds number. Due to the use of the carbon black marker, the polyol reactant laminae appear dark in the captured images. The striation thickness distribution was measured in a polymer sample extracted from the center of the tube mold to avoid the effect of further striation thinning in the shear flow in the tube. Large striations ($500 \mu\text{m}$) were not recorded since they correspond to unreacted polymer, and, due to optical

limitations, striation thickness less than 10 μm were not detectable. The authors concluded that the obtained average striation thickness is not quantitatively accurate to validate any of the proposed models at the time. Stoichiometry seems to have been the only concern for setting the two feeding streams flow rates, resulting in $r_s = 1.5$, which presents some concerns since the jets' imbalance can affect strongly mixing quality.

Baldyga and Bourne (1983) developed a model that estimates a distribution of striation thickness. The proposed model also predicts a reduction of scales in the light of statistical theory of turbulent diffusion combined with an exponential residence time distribution of the fluid inside the mixing zone of the chamber to provide a distribution of lamellae thickness. Further deformation of the striation scales below the Kolmogorov scale, where viscous forces and a dependence on Re would be significant, was not considered. Most certainly due to this assumption, results from this model show good agreement with the experimental results of Kolodziej et al. (1982), where scales smaller than 10 μm could not be detected. Later, Bourne and Garcia-Rosas (1985) included in the model of Baldyga and Bourne (1983) the further striation thinning due to laminar parallel flow in the remain mixing chamber. The model shows that the average striation thickness is reduced at least one order of magnitude in the length from the dynamic mixing zone to the outlet. The striation thickness probability distribution below or equal to 10 μm also increases significantly from zero to more than half. These results seemed to explain, only from isotropic and homogeneous turbulence theory considerations of Tucker III and Suh (1980) and Baldyga and Bourne (1983), how mixing length scales are reduced in the RIM machine's mixing chamber to values that allow complete chemical reaction in the mold, despite of non turbulent flow in the mixing chamber in the range of industrial practice.

May (1996) obtained a striation thickness distribution in RIM from a computational fluid dynamics point of view. A field equation was derived from statistical theory of turbulent diffusion in order to describe the striation thickness reduction. May (1996) presents similar results to the ones obtained by Baldyga and Bourne (1983). The difference between them reside in that no assumption for the chamber residence time distribution is necessary by solving directly the flow field and turbulent properties equations.

Since May (1996), no known further developments have been published on the characterization of the lamellar structure in RIM. In the computational study of mixing in RIM made by Santos et al. (2005) the diffusivity value was set to a value too high to be able to identify any lamellar structures from the results. In the CFD study of Fonte et al. (2011), despite avoiding the effect of diffusion, the spatial discretization was not sufficiently refined to include all scales. Even with the evolution of the available computational power, the description of the reduction of scales in RIM by direct simulation of material surfaces is still extremely resource consuming. Even for a simplified 2D model of the flow, the exponential generation of interfacial area demands the use of an increasingly dense spatial discretization in time to properly describe the interfaces and avoid numerical problems. Up to this date, the description of a lamellar structure in RIM from simulation of fluid interfaces is yet not feasible for 3D flows. Instead, the length scales generated in RIM can be predicted by the stretching distributions in the flow since their statistical properties are identical (Paul et al., 2003).

The problem of maintaining the mixing regime during RIM operation or other Confined Impinging Jets Reactors (CIJR) applications has also been the object of study of several authors. Malguarnera and Suh (1977) were the first to show experimentally with a RIM machine prototype that, in addition to satisfying the desired mass flow rate ratio, set by stoichiometry, the momentum ratio of the fluid components must be equal to 1. Malguarnera and Suh (1977) proposed that when a volumetric stoichiometric ratio different from 1 is desired, it is necessary to use different section area injectors.

Sebastian and Boukobbal (1986) studied experimentally the effect of the jets momentum ratio on the actual polymerization. Their study has shown that a higher adiabatic temperature raise, which is directly linked to product yield and mixing efficiency in exothermic reactions, is obtained when the momentum ratio is closer to one.

Hosseinalipour and Mujumdar (1997) studied numerically the effect of unequal momenta jets impinging in two-dimensional confined opposing reactor in steady laminar regime. In this work the inequality in the jets' momenta is achieved using equal or unequal widths of the two dimensional jets by adjusting the average velocity appropriately, showing the effect in the flow and its thermal characteristics. Johnson (2000) observed a significant alteration of the flow field under unequal flow rate conditions in RIM from numerical flow simulations and experimentally for an equal injectors diameter mixing chamber. Changing the flow rate of one injector moves the impingement point towards the lower momentum rate jet, stabilizing the flow field to some extent. Johnson (2000) has shown the formation of large areas of recirculation in the mixing chamber that lead to poor mixing due to the possibility of unmixed fluid leaving the chamber and the increase in the residence time of the mixing fluids.

Santos et al. (2002) and Santos et al. (2010) have also shown from Laser Doppler Anemometry (LDA) and computational simulations of the flow that the momentum ratio between the jets is a critical parameter for the dynamic behavior of the system where the jets oscillations were only observable on a very narrow interval of the momentum ratio. This work has also shown that, by changing the injector diameters, a sustainable chaotic flow regime can be obtained for flow rate ratios different from the unity as long as the diameter change imposes a unitary value for the momentum ratio.

Siddiqui et al. (2009) also studied numerically and experimentally the effect of unequal flow on the average energy dissipation in a CIJR, showing a sharp decrease of energy dissipation in a CIJR, as the difference in flow rates increases, to approximately 40% of the value for equal flow rates for lower flow rates ($Re \sim 2100$). For higher flow rates $Re \geq 3500$, Siddiqui et al. (2009) reports a transition to a turbulent flow regime stabilizing the decrease of energy dissipation with the increase of flow rate difference. Under these conditions, energy dissipation retained 84% of the value in balanced flow all the way to a 30% difference in flow rates, suggesting that reservations about the practicality of operating CIJRs under non-ideal plant conditions and varying inlet flow rates can be set aside. Despite these results, the range of industrial interest for RIM operation goes only to Reynolds numbers up to 600, as high monomers' viscosities would imply an excessive use of pumping energy for higher Re , making these recommendations not applicable.

Steffen Schütz et al. (2009) studied the impact of the injectors' viscosity ratio in the flow, and observed that the jets' impingement point position is shifted from the center towards the injector where the higher viscous fluid is injected due to the increased frictional forces acting on the higher viscous jet, leading to a quicker jet expansion. They have also observed that an asymmetric velocity field results in a worse distribution of shear and elongation forces and in an incomplete usage of the mixing volume.

More recently, Nunes et al. (2012) for RIM and (Krupa et al., 2012) for T-shaped CIJRs have also shown experimentally with consecutive-competitive reactions, specifically used in micromixing characterization, that mixing quality decreases when the flow rate ratio is different from unity.

Main results and Conclusions

The main results of this dissertation are the definition of the flow regimes observed in the range of Reynolds number of industrial interest for the production of polymers from RIM ($50 < Re < 600$), the quantification of the stretching and striation thickness distribution in the flow, and the conclusions about the impact of the jets' unbalancing on the flow. Three different flow regimes were identified:

- $Re < 105$: The flow is laminar and steady with poor mixing properties. The flow remains segregated with each side of the mixing chamber containing mainly one of the injected fluids.
- $105 \leq Re \leq 110$: A dynamic flow regime is onset. The flow evolves to a self-sustained oscillatory periodic laminar regime characterized by the disturbance of the two impinging jets' interface and the formation of alternated detaching vortices in the mixing chamber downstream the inlet.
- $110 < Re < 600$: The formation of the vortices occurs earlier in the flow, i.e. closer to the impingement point, and start to interact with the inlet jets. At this point, the flow develops to a self-sustained laminar chaotic regime with strong dynamics and good mixing properties.

Results have shown that advective mixing in opposed impinging jets is observed to occur above a critical Reynolds number $Re_c > 110$. This critical Reynolds number value is shown from PIV experiments to be same for two different mixing chambers with different injectors' diameters.

From the calculation of the stretching experienced by the advected passive elements in the flow (see Figure 2a), this work shows that, in the mixing regime, the reduction of scales in opposed impinging jets mixers results from an increase of the stretching rate combined with a strong flow reorientation, both at the top of the mixing chamber. In reactive polymer injection, the combination of the two mechanisms (stretching and reorientation) allows the flow to exponentially generate interfacial area between the two monomers or pre-polymers in RIM and reduce scales to a value that avoids diffusional limitations during the polymerization reaction in the mold. In this regime the flow is shown to produce nearly exponential average stretching of fluid elements with time, a feature of chaotic flows. Impingement mixing is observed to occur in the top of the chamber until a distance approximately equal to twice the chamber diameter from the top wall. In the remaining chamber the flow develops a nearly parallel profile. The flow in the bottom of the chamber still contributes to further reduction of mixing length scales but linearly in time due to the orientation of trajectories with the streamlines. Further reduction of striation thicknesses in the monomers or pre-polymers mixture is observed with the increase in Reynolds number.

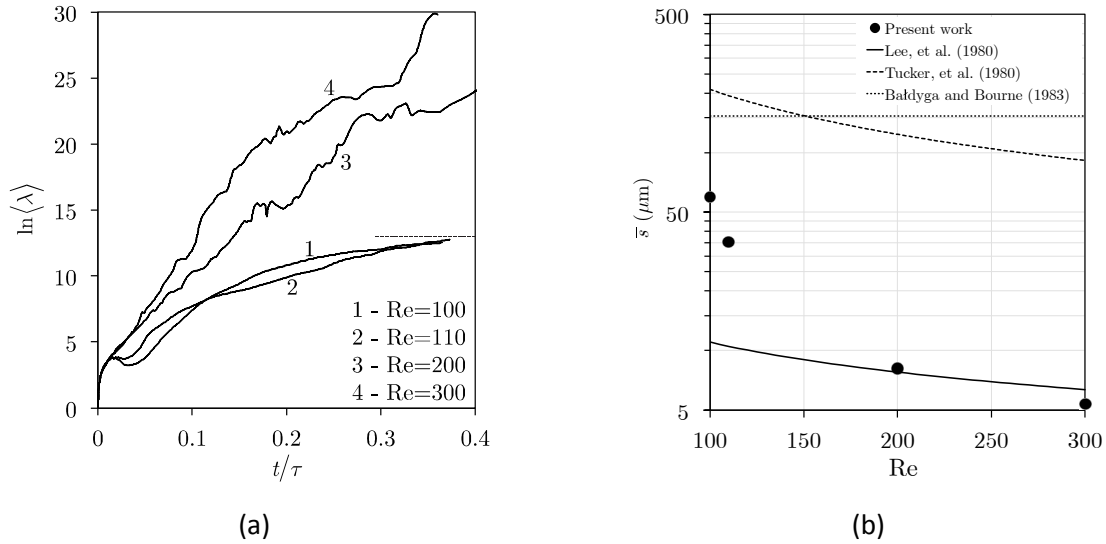


Figure 2. a) Average particle stretching at several Reynolds numbers as a function of time; b) Mean striation thickness as a function of Reynolds number

Computational and experimental results of this work are compared to theoretical models existent in the literature for the estimation of laminae scales in impingement jets mixing (see Figure 2b). Results show to be consistent with the kinematic model of Lee et al. (1980), rather than with models based on the statistical theory of turbulent diffusion (Tucker III and Suh, 1980; Baldyga and Bourne, 1983; Bourne and Garcia-Rosas, 1985; May, 1996). The observed patterns and distribution of lamellae thicknesses indicate that the flow up to $Re = 600$ and for $Re \geq Re_c$ is laminar and that mixing is achieved by the advective mechanisms of stretching and folding. The results of this dissertation strongly indicate that isotropic and homogeneous turbulence considerations for the modeling of mixing scales do not provide a correct physical description of the flow.

CFD Simulations and PLIF experimental results show as well the formation of striation scales thin enough to be faded by diffusion during time periods close to the mixing chamber mean residence time. This result suggests an additional mechanism of *rupturing*, associated with stretching and folding mechanisms, capable of generating isolated blobs of material in the medium.

Unbalanced jet conditions were found to affect the flow significantly by moving the impingement point towards the chamber walls. Under unbalanced conditions, the jets' oscillations are partially or completely damped, even when a dynamic flow regime is expected. PLIF and PIV results for different jets' mass flow rate ratios show the importance of maintaining the jets balanced, which corresponds to an average impingement point of the two jets in the center of the chamber (see Figure 3). When the jets' impingement point is slightly deviated from the center of the mixing chamber, by increasing or decreasing one of the jets' flow rate, the mixing degree of the two feeding streams decreases significantly and becomes more variant in time. This behavior is shown to occur even when the deviation of the jets' impingement point position from the center of the chamber corresponds to increasing the total kinetic energy rate of one of the streams. This suggests that, even by increasing the energy dissipated in the flow by shear, by increasing one of the injector's flow rate, the flow field is less efficient in the reorientation of striations in the main direction of deformation and in reduction of scales by eddy engulfment. The results show that the operation of CIJs mixers/reactors, in the laminar regime, is not only focused on the amount of energy supplied to the system for dissipation, but also in maintaining and maximizing the natural chaotic oscillation of the two impinging jets.

Contrarily to the reported by other authors (Malguarnera and Suh, 1977; Sebastian and Boukobbal, 1986; Hosseinalipour and Mujumdar, 1997), optimum conditions are obtained when the kinetic energy rate of both jets are balanced, and not the respective momenta.

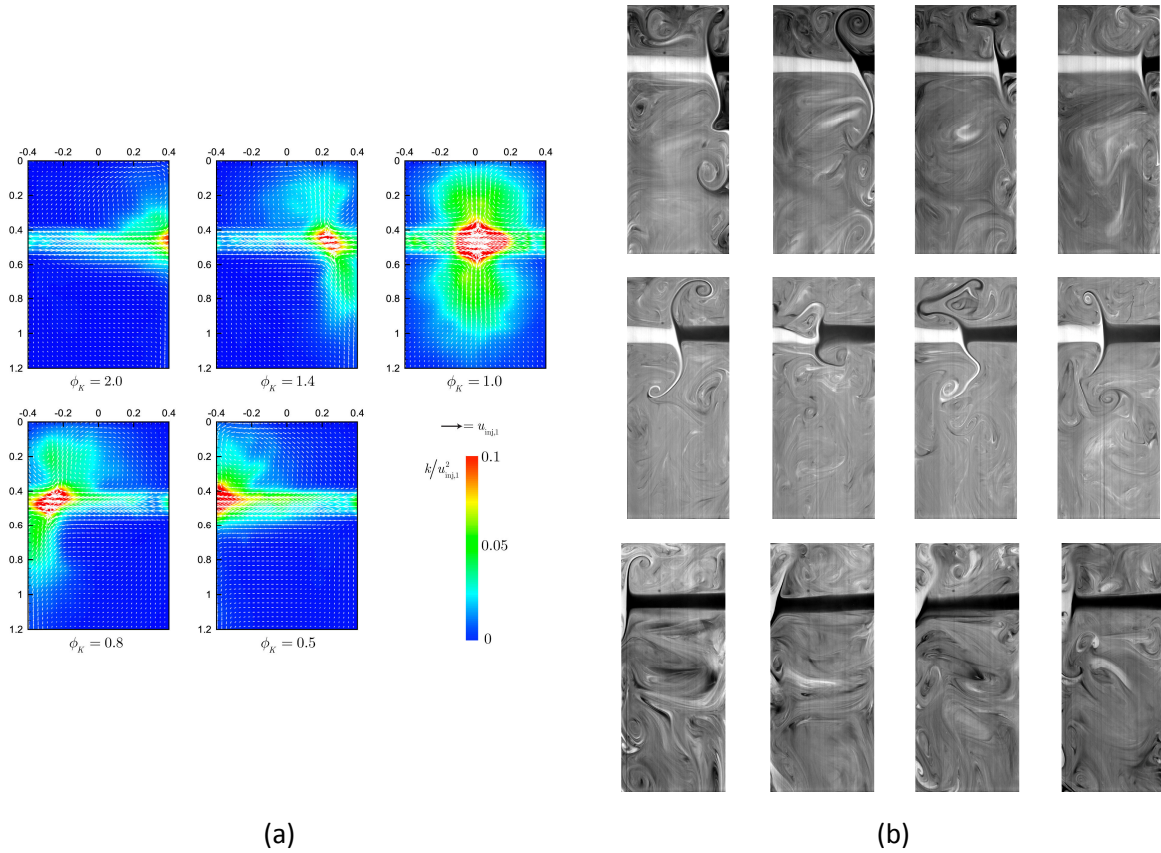


Figure 3. Effect of the position of the impingement point on mixing quality at $Re=300$: a) PIV Velocity field vector maps and non-dimensional velocity fluctuations contours as a function of the kinetic energy rate ratio; b) Instantaneous concentration fields obtained from PLIF experiments.

When the Reynolds number is increased, maintaining the jets' kinetic energy rate ratio and at values different from one, the impingement point moves towards the chamber walls, closer to the lowest Reynolds number jet side. This phenomenon indicates that the impinging jets flow becomes more sensitive to small deviations in flow rates as Re is increased. This shows that during in RIM operation the increase of Re is reflected in the need for more precise and strict monomers or pre-polymers flow rate control and the system becomes more sensitive to temperature of the polymer production plant.

The present dissertations clearly states that impingement mixing must occur at the center of the mixing chamber for better mixing efficiency, setting the kinetic energy rate ratio as estimated by the proposed elastic analogue model, and for $Re > 110$. The injectors' diameter of the mixer can be adapted to comply simultaneously with reaction stoichiometry. The results provide deeper understanding on the nature of mixing by CIJs in the laminar regime, with particular interest on the optimization of mixing of high viscosity liquids, as in the RIM process.

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