Methods for the characterization and comparison of mixing efficiency of different confined opposing jet mixing devices.

M. Bertrand, E. Plasari, P. Baron

METHODS FOR THE CHARACTERIZATION AND COMPARISON OF MIXING EFFICIENCY OF DIFFERENT CONFINED OPPOSING JET MIXING DEVICES

M. Bertrand-Andrieu[a]*, E. Plasari[b], P. Baron[a]

[a] *Commissariat à l'Energie Atomique, C.E.A Valrhô, SCPS, Bâtiment 399, BP 17171, 30207 Bagnols-sur-Cèze, France ;* murielle.bertrand@cea.fr

[b] *Laboratoire des Sciences du Génie Chimique - CNRS, Ecole Nationale Supérieure des Industries Chimiques - INPL, 1 rue Grandville, B.P. 451, 54001 Nancy Cedex, France.*

Abstract. The work reported here describes the methods to characterize and compare mixing efficiency of three high-intensity mixers of the same family : T-shaped tube, Y-shaped tube with a 90° angle and Hartridge-Roughton mixing device. Mixing efficiency is investigated using two chemical methods, the "iodide-iodate" micromixing test reaction and an acid-base neutralization. Experiments are carried out under the same operating conditions, with Reynolds numbers from 15,000 to 40,000. Micromixing time constant values determined by both methods are very close and lead to the same mixer classification. Under the same operating conditions, the Y-tube mixer has the lowest mixing efficiency, while the Hartridge-Roughton mixing device is much more efficient than the two other mixers.

Key words: Mixing efficiency, T- tube, Y- tube, Hartridge-Roughton mixing device, Micromixing test, Neutralization test.

1. INTRODUCTION

Many chemical reactions are fast relative to mixing so that conversion occurs before the reagent homogenisation is achieved. In such cases, concentration gradients affect considerably the final product properties. For precipitation systems, the particle size distribution is well known to depend on the supersaturation levels governed by the mixing process [1]. When liquids are very viscous (polymer production, ceramic, glassware and food processing), mixing may influence the selectivity and rate of chemical reactions and the molecular weight distribution. Therefore, when reaction and mixing proceed simultaneously and not consecutively, the way of contacting reagents appears to be a crucial parameter in the control of the final product quality. Thus, a particular attention should be paid to the choice of the mixing device and information about mixing characteristics is of considerable interest for the reactor design. That is why, the work reported here describes two methods to characterize and compare mixing efficiency of confined opposing jet mixing devices, which are commonly used to contact rapidly and continuously two fluid streams. These methods have been applied to three high-intensity mixers of the same family: T-shaped tube, Y-shaped tube with a 90° angle and Hartridge-Roughton mixing device [2,3] in which the jets enter tangentially into a circular chamber. These mixing devices are presented in Figure 1. The objective of this document is to provided rapid and easy to implement experimental methods avoiding long studies.
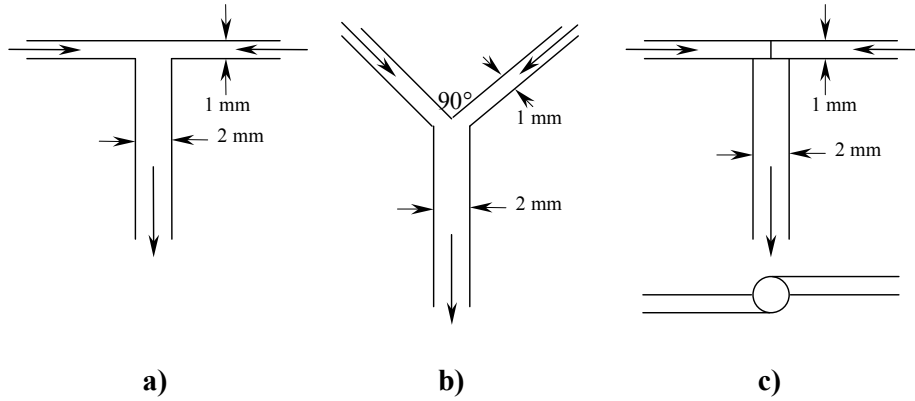
Figure 1: Schematic presentation of high-intensity confined opposing jet mixing devices: a) T-tube, b) Y-tube (90° angle) and c) Hartridge-Roughton mixing chamber.

Many studies on mixing efficiency are available in the literature for some of these devices [4-10]. However, tests being performed in different size and geometry and under different operating conditions, it is not possible to point out the most effective system. That is why, in order to respond to this question, we have studied the mixing intensity in respective tubes of the same dimension, only the geometry of the impinging fluid streams being different as shown in Figure 1.

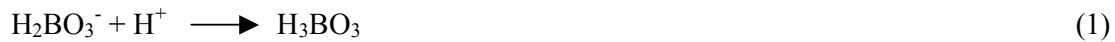## 2. MIXING EFFICIENCY CHARACTERIZATION

Macromixing involves turbulence which rapidly decreases the segregation scale down to the Kolmogoroff velocity microscale (λ_k), and then the molecular diffusion process achieves the complete homogenisation. Micromixing in the three devices studied (Figure 1) is investigated using two chemical methods. The first one consists in a micromixing test based on the competing reactions which keep in memory the mixing efficiency through the product distribution. On the contrary, the second method is based on an acid-base neutralization reaction where the mixing efficiency is characterized using the reaction volume measured by visualization. Experiments are carried out under the same operating conditions; the fluid velocity in the mixing tube varies from 8 to 20 m s^{-1}, with a Reynolds number of the order from 15,000 to 40,000.

2.1 Micromixing test reaction

Among the several chemical tests of micromixing characterization available in the literature, we select the well known "iodide-iodate" method developed by Villermaux and co-workers [11], due to its flexibility and its ease of processing [12].

Iodide- iodate method
The "iodide-iodate" method is based on a system of two parallel-competing reactions. The first one is a "quasi-instantaneous" neutralization and the second one is the classical Dushman's oxydo-reduction reaction. The whole reaction system may be written as follows:

$$H_2BO_3^- + H^+ \longrightarrow H_3BO_3 \tag{1}$$
$$5I^- + IO3^- + 6H^+ \longrightarrow 3I_2 + 3H_2O \tag{2}$$
$$I^- + I_2 \longrightarrow I_3^- \tag{3}$$

The test consists in contacting a strong acid (sulphuric acid) with a iodide, iodate and borate ion mixture. For a perfect mixing, the injected acid reacts instantaneously with borate ions according to the neutralization reaction infinitely faster than reaction (2). On the contrary, in the presence of a local acid excess, iodine is formed and the I_3^- concentration, measured by spectrophotometric absorption, allows to quantify the segregation intensity through a segregation index X_S. X_S varies between 0 for a perfect micromixing state and 1 for a total segregation one:

$$X_S \quad = \quad \frac{n_{I_2} + n_{I_3^-}}{n_{H^+,0}} \left[2 + \frac{n_{H_2BO_3^-,0}}{3\, n_{IO_3^-,0}} \right] \tag{4}$$

where n_i is the mole number and subscript 0 represents the initial conditions.

The experimental procedure described by the authors relates to a batch stirred tank in which a small acid volume is slowly injected. As our experimental conditions require to operate with equal volumes in continuous mode, we adapt the method by using a low acid concentration. In these conditions, acid aggregates are flooded in the iodide, iodate and borate mixture and can be supposed independent from each other. According to Baldyga and Bourne [13], for a Schmidt number lower than 4000, micromixing process is supposed to be controlled by the engulfment step. Our conditions leading to a Schmidt number of about 1000, the experimental results are interpreted using the Incorporation Model [14] where the acid aggregates are assumed to gradually grow by engulfment of surrounding fluid. Considering an exponential incorporation function, the experimental segregation indexes can be associated with the characteristic micromixing time constant t_m.

Experimental
Experiments are carried out in a specific apparatus developed by the Commissariat à l'Energie Atomique and the Chemical Engineering Science Laboratory at Nancy [15]. The operating principle is schematically shown in Figure 2 and a photo is given in Figure 3.
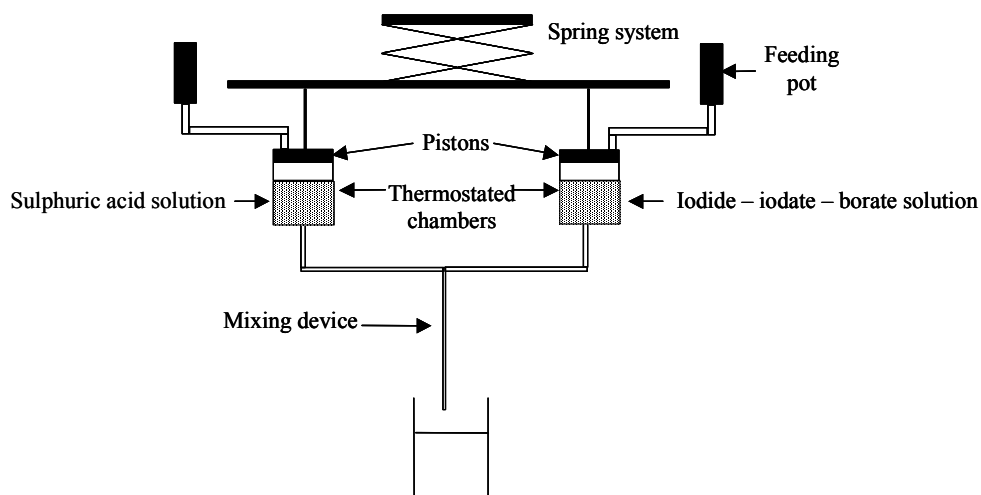

Figure 2: Schematic diagram of the experimental set-up for iodide-iodate tests.
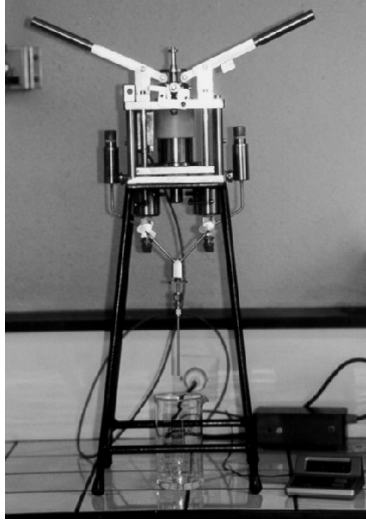
Figure 3: Micromixing test apparatus.

The mixing device is connected to two thermostated chambers equipped with pistons moved by a spring mechanism. The iodide, iodate and borate ion mixture and the sulphuric acid solution, initially contained into the chambers, are simultaneously forced into the mixing device by the piston release. The final mixture recovered at the tube outlet is rapidly analysed by spectrophotometric absorption at 353 nm to determine iodine concentration.
The initial reagent concentrations are as follows:

$[I_2]_0 = 3 \ [IO_3^-]_0 = \frac{3}{5} \ [I^-]_0 = 0.021 \ mol/L^{-1}$

$[H_3BO_3]_0 = 0.5 \ mol/L^{-1}$
$[NaOH]_0 = 0.25 \ mol/L^{-1}$
$[H_2SO_4]_0 = 0.05 \ mol/L^{-1}$

2.2 Neutralization tests

The devices are made of glass, so mixing characteristics can be investigated using a neutralization reaction between hydrochloric acid and sodium hydroxide solution in the presence of bromotymol blue as indicator. The initial H^+ concentration C_{A0} is taken 5 % higher than the initial OH^- concentration C_{B0} in order to accomplish the neutralization of hydroxide ions. The yellow color corresponds to the total neutralization, so to a total homogenization at molecular scale of reacting fluids, whereas the blue one indicates a local concentration inhomogeneity.

Theory
Using the Incorporation Model in the case of instantaneous reactions, a simple relationship can be obtained between the decolourization time "t_{dec}" and the micromixing time constant t_m. According to the model, the basic aggregates are assumed to gradually incorporate the surrounding acid fluid and to grow as an exponential time function:

$$V_B = V_{B0} \ exp\left(\frac{t}{t_m}\right) \qquad (5)$$

At decolourization time t_{dec}, the acid mole number incorporated is equal to the base mole number initially present in the basic aggregates, so:

$$V_{iA}\left(t_{dec}\right) = \frac{V_{B0}\, C_{B0}}{C_{A0}} \tag{6}$$

where V_{iA} is the acid volume incorporated and t_{dec} is the mixing time defined as follows:

$$t_{dec} = \frac{L_{dec}}{u} \tag{7}$$

u being the flow velocity in the tube and L_{dec} corresponding to the length necessary for the neutralization (the blue length).

Consequently, at decolorization time, the basic aggregate volume has grown as:

$$V_B(t_{dec}) = V_{B0} + V_{iA}(t_{dec}) = V_{B0}\left(1 + \frac{C_{B0}}{C_{A0}}\right) \tag{8}$$

From (5) and (8), we obtain:

$$t_m = \frac{t_{dec}}{\ln\left(1 + \dfrac{C_{B0}}{C_{A0}}\right)} \tag{9}$$

Knowing that $C_{B0}/C_{A0} \approx 1$, we obtain the following relation between the decolourization time and micromixing time constant:

$$t_m \approx 1.44\, t_{dec} \tag{10}$$

Experimental
Experiments are performed continuously in transparent mixers made of glass and connected to 100 L feeding tanks, as shown in Figure 4 [16].
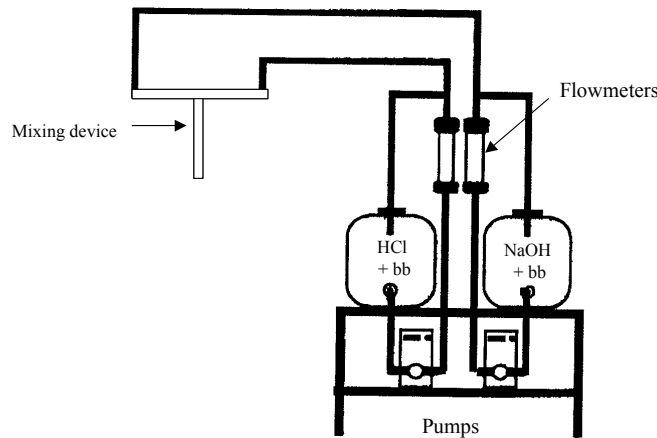

Figure 4: Experimental set-up for neutralization tests.

Reagents, containing initially blue bromothymol in order to prevent the decolourization from the dilution effect, are supplied by pumps with a 25 mL.s^{-1} flow rate.

The decolourization time can be easily calculated from visual observations by measuring the tube length coloured in blue.

Table 1. Micromixing time constant t_m (in ms) determined by two methods (fluid velocities in the central tube of the order of 8 m.s^{-1}).

Mixer type	Iodide/iodate method		Neutralization method	
	X_S x10^3	t_m (ms)	t_{dec} (ms)	t_m (ms)
T-tube	1.8 ± 0.2	3.3 ± 0.4	2.0 ± 0.4	3.0 ± 0.6
Y-tube	2.4 ± 0.2	4.9 ± 0.7	2.9 ± 0.6	4.3 ± 0.8
Hartridge-Roughton mixing device	0.9 ± 0.1	1.7 ± 0.2	0.8 ± 0.2	1.2 ± 0.2

3. RESULTS AND DISCUSSION

For the iodide-iodate method, experimental runs show that the micromixing time is estimated with a 15% error, whereas the neutralization tests lead to an error estimation of about 20 % due to the experimental appraisal of the decolorization length. Table 1 compares micromixing times obtained by both methods, iodide-iodate test and acid-base neutralization, in the case of a fluid velocity in the central tube of the order of 8 m.s^{-1}. The micromixing time constant values are very close and lead to the same mixer classification. The use of the iodide/iodate method being much more complicated than the acid-base neutralization, we recommend the second method in all cases when the experimental conditions of fluid mixing visualization are fulfilled.
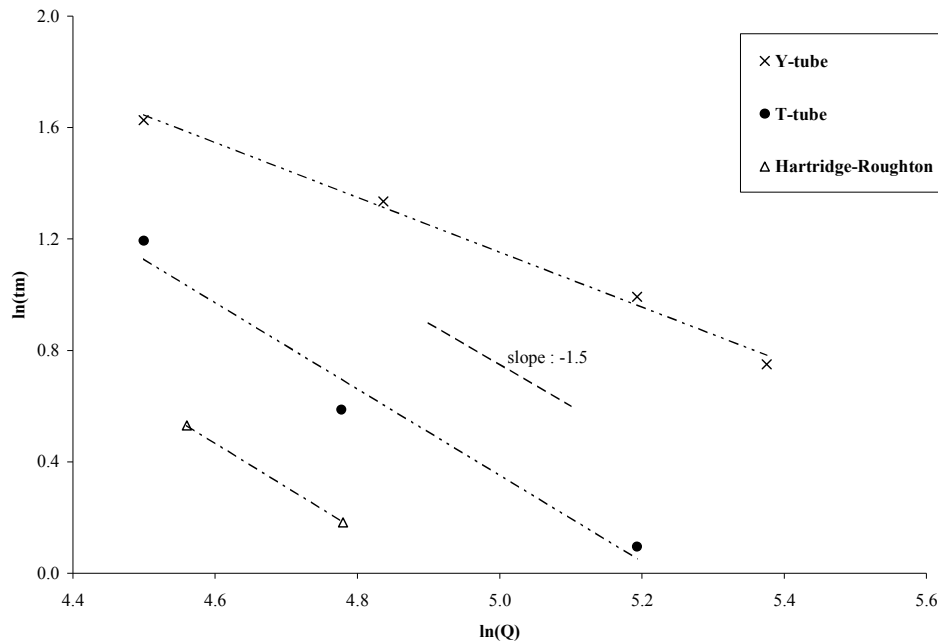

Figure 5: Flow rate influence on the micromixing time – Iodide-iodate method.

For the iodide-iodate test, the segregation index X_S and the micromixing time constant t_m are determined for different reagent flow rates. Figure 5 presents a logarithmic plot of micromixing time constant (in ms) against flow rate (in $L.h^{-1}$). This figure clearly shows that the Y-tube mixer has the lowest mixing efficiency, while the Hartridge-Roughton mixing device is much more efficient than the two other mixers. In addition, its geometry is simple and the mixer can be easily produced. For these reasons, between the confined opposing jets mixing devices studied in our work, we recommend to use the Hartridge-Roughton mixing device.

In the case of both T-tube and Hartridge-Roughton device, the micromixing time constant decreases with the flowrate according to the formula:

$$t_m = K' \, Q^{-1.55} \tag{11}$$

This result is in good agreement with the Incorporation Model where:

$$t_m = K \sqrt{\frac{\nu}{\varepsilon}} \tag{12}$$

Knowing that, in a smooth tube, the energy dissipation rate is proportional to u^3, the micromixing time varies as $Q^{-3/2}$. The same result is obtained by Johnson and Prud'homme [17] who have studied micromixing in confined impinging jets. On the contrary, data obtained for the Y-shaped tube lead to a lower slope value equal to 1.1. This is probably due to the influence of the Y-tube entry on the fluid velocity distribution in the upper part of the central tube giving a law of the energy dissipation rate against velocity with an exponent lower than 3.

4. CONCLUSION

The iodide-iodate method, based on the competing reactions which keep in memory the mixing progress through the product distribution, and the decolourization method, based on the measurement of the reaction volume, lead to micromixing time constant values very close to each other and can successfully characterize the mixing efficiency of rapid mixing devices. Both methods described here make it possible to compare mixing efficiency of the T-tube, Y-tube and Hartridge-Roughton mixing device and to propose a mixer classification. Nevertheless, the second method is much simpler for use (simple experimental investigation, very simple treatment of experimental data), so we recommend it in all cases when the experimental conditions of fluid mixing visualization are fulfilled.

The three mixers studied are characterized by very low segregation indexes with micromixing time constants of few milliseconds. Nevertheless, a sensible difference in mixing efficiency exists between them. The highest mixing efficiency is obtained for Hartridge-Roughton geometry. In addition, its geometry is very simple to be realized and used. For these reasons, we recommend as the first choice the Hartridge-Roughton mixing device.

NOMENCLATURE

C_A : acid concentration ($mol.m^{-3}$)
C_B : base concentration ($mol.m^{-3}$)
K : constant (-)
L_{dec} : decolourization length (m)
n_i : mole number of element I
Q : flowrate ($m^3.s^{-1}$)
t_{dec} : decolourization time (s)

t_m : micromixing time constant (s)
u : flow velocity ($m.s^{-1}$)
V_B : basic aggregate volume (m^3)
V_{iA} : acid volume incorporated (m^3)
Xs : segregation index (-)
ν : kinematic viscosity ($m^2.s^{-1}$)
ε : local energy dissipation rate ($W.kg^{-1}$)

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