A New Mathematical Model for the Prediction of Internal Recirculation in Impinging Streams Reactors

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ABSTRACT: *A mathematical model for the prediction of internal recirculation of complex impinging stream reactors has been presented. The model constitutes of a repetition of a series of ideal plug flow reactors and CSTR reactors with recirculation. The simplicity of the repeating motif allows for the derivation of an algebraic relation of the whole system using the Laplace transform. An impinging streams reactor system with one axial and two tangential inlet fluid streams was constructed and considered as a case study. The model predicts satisfactorily the complex and flow rate dependent experimental residence time distribution functions obtained employing a pulse tracer method for different total flow rates of the incoming feed. The variation of the controlling parameters with changing the total inlet flow rate are discussed. The presented model can predict complex internal recirculation streams within the impinging streams reactor system.*

KEYWORDS *Impinging streams reactor; Residence time distribution; Internal recirculation.*

INTRODUCTION

The impinging streams were introduced as a novel technical method in 1961 in order to intensify mass and heat transfer between phases [1]. In such a system, two or more streams flow parallel or countercurrent to each other and collide in an impingement zone. This collision results in high relative velocity between phases, enormous turbulence, internal recirculation, and oscillation motion of particles [2-8]. This subject has attracted the interest of the academic world and during the last decades, several different types of devices that apply impinging streams have been developed [9-10]. Some of these are currently successfully used in industrial plants such as entrained-flow gasifiers with opposed multi-burner [11] and Trost Jet miller [12].

In designing a reactor and predicting its performance, it is very effective to determine the Residence Time Distribution (RTD) of the fluid which flows through that reactor. The length of time that various elements of the exiting fluid spend within the reaction system maybe determined by experimentally measuring the RTD [13,14]. It may also be used for determining the flow pattern within the reactor which is very important

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in the up-scaling of laboratory scale experiments to industrial scale [15-16]. Simulating a non-ideal system by a configuration of ideal systems consisting of ideal perfect plug or ideal perfect mixed reactors or a combination of these two types of ideal reactors is also possible by measuring the RTD [17].

Sohrabi and his co-workers have applied various impinging stream reactors in different processes and reported the residence time distribution of them during the last two decades [5, 14, 18-33]. Among these reactors, there is an axial-tangential reactor which was applied to intensify enzyme reactions, photodegradation of 4-nitrophenol and *p*-nitrophenol [31-33]. The fluctuations in RTD curves which is reported for this type of reactor [31] may not be predicted by the common RTD models including tank in series with equal-sized stirred tanks, [19] tank in series with descending-sized stirred tanks [20], Gamma distribution with bypass (GDB) [13,31], direct simulation Monte Carlo (DSMC) method [22] and Markov-chains discrete time model [18,34].

These models are good in predicting the RTD curves for common configurations of impinging stream reactors, but axial-tangential impingement ones are prone to result in significant internal recirculation of material within the reactor volume, according to RTD curves reported here. None of the cited models have been investigated for the prediction of internal recirculation effects. Mathematically, only the Markov-chain model may result in limited recirculation effects. Therefore, these models are not adequately consistent with the physics of impinging streams. In this research, RTD curves for an axial-tangential impinging streams reactor are experimentally determined and a mathematical model is presented attempting to predict the complex RTD curves observed using different flow rates.

EXPERIMENTAL SECTION

Materials and analytical procedure

RTD experiments were performed using distilled water as working fluid. The tracer used in this study has been reagent grade methyl orange, which was purchased from Sigma-Aldrich. The RTD curve was obtained by imparting a pulse input of the tracer material to the inlet streams simultaneously, using equal amount of material for each stream. Measurement of the tracer concentration in the outlet stream was preformed through sampling and spectrophotometric analysis. The latter analysis was performed using a UV-visible spectrometer (Hatch/Lange (DR/2800)) at a wavelength of 507 nm using a calibration curve.

Impinging stream reactor design and operation

The impinging stream reactor used in this study is a cylindrical vessel made of Plexiglas. The vessel diameter is 7 cm and its height is 4 cm. Two nozzles are perfectly connected tangentially to the surface of the vessel as shown in Fig. 1a. Nozzles have the same internal diameter (1 mm) resulting in a same jet velocity for both of them. The third nozzle is connected vertically beneath the vessel. The latter and the outlet nozzle have a similar cylindrical geometry (1 cm in height and 1 cm in diameter).

In Fig. 2, a schematic of the experimental apparatus is presented. It consists of a fluid injection system (water tank, pumps, valves and rotameters), the impinging stream reactor and a rotating disk connected to a power supplier which was used for sampling at regular time intervals. The disk was immediately located under the discharge port of the reactor.

Flow rate of the jet streams was controlled by valves and measured by rotameters before entering the reactor at different operating conditions (different flow rates).

During each run, the disk is made to rotate at a desired speed. The rotating speed of that was controlled by the electrical power of a supplier. By changing the speed of rotation, different intervals of sampling were obtained. The disk consisted of several cells and used to collect samples of the outlet flow of the reactor at regular time intervals.

RTD experiments were performed at different injection flow rates of impinging streams in order to get an insight into the flow pattern of the fluid elements in the reactor volume and to determine the effect of flow rate on the flow pattern. To study the effect of flow rate on the RTD, a range of flow rates (0.25, 1.0, 1.5 and 2 L/ m) were applied for the injected flow via three nozzles into the reactor vessel. The flow rate of the axial inlet nozzle had been half of the total flow rate for each experiment. The flow rate of the tangential nozzles was equal. RTD determination was performed applying a pulse injection of tracer.

In these experiments, water was fed to the reactor as working fluid and methyl orange (1000 ppm) as tracer.

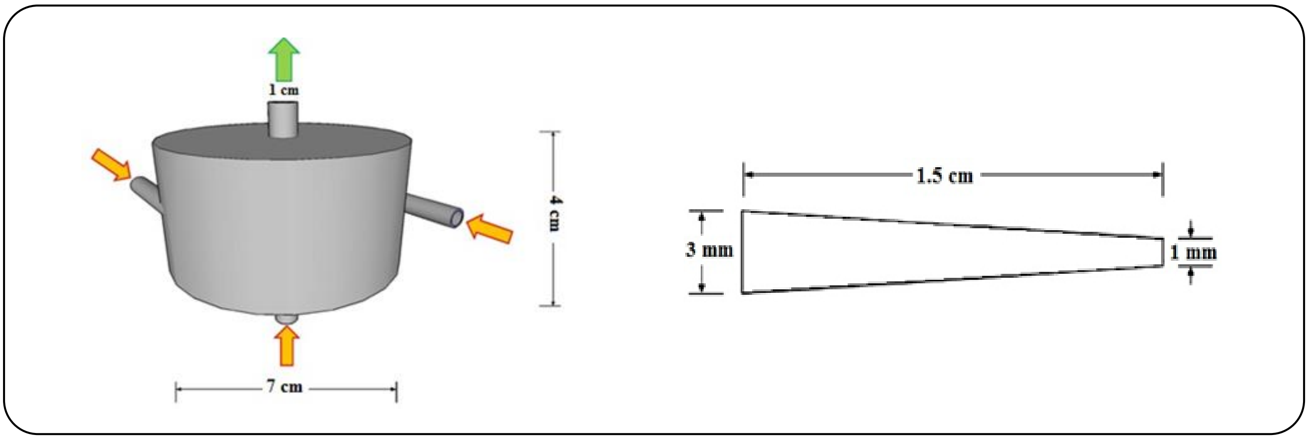

Fig. 1: a) Impinging streams configuration. b) Tangentially installed nozzle's geometry.

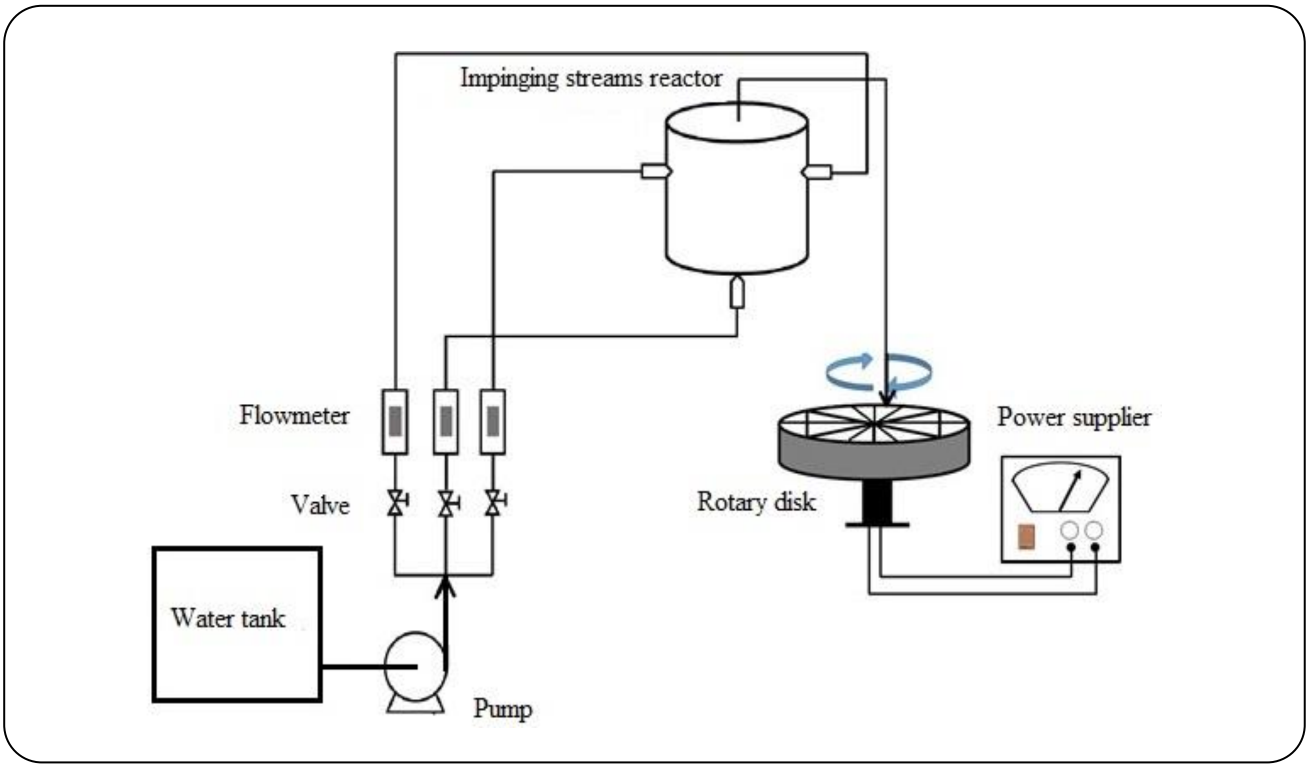

Fig. 2: Schematic illustration of the experimental apparatus.

At time t=0, tracer was injected instantly and instantaneously into the streams prior to entrance of nozzles, using three similar syringes. The disturbance to the bulk flow caused by the injection of the methyl orange may be assumed to be negligible. Samples were collected at the outlet of the reactor at regular time intervals (Fig. 2). A UV-visible spectrophotometer was used to analyze the collected samples and to determine the concentration of injected methyl orange.

In order to review the main parameters of conventional theoretical models of RTD, a brief review of them is given in the following section.

THEORETICAL SECTION

Brief review of theoretical models of RTD

The tank-in-series model has been conventionally used by researchers to describe the RTD of simple impinging streams reactor systems [19]. The actual volume of the reactor is considered to be consisted of N equal-size ideal CSTRs in this model. The number of stirred tanks is the only parameter considered by this simple model in order to describe the fluid behavior. The RTD equation takes the following form [35]:

$$E(\theta) = \frac{N(N\theta)^{(N-1)}}{(N-1)!} E^{-N\theta} \tag{1}$$

Where θ is the dimensionless time and $E(\theta)$ the dimensionless exit stream time distribution function. Based on Equation (1), the RTD function takes the shape of a mono-modal curve. This is while recirculation generally may result in multi-modal RTD curves.

The GDB model has also been used to describe the RTD of impinging stream reactors [21]. This model consists of a gamma mixing section followed by an ideal plug flow reactor. The RTD equation in this case is as follows:

$$E(\theta) = \frac{\beta(\alpha\beta)^{(\alpha)}}{(1-\beta x)\Gamma(\alpha)} \beta(\theta-1)^{\alpha-1} \tag{2}$$

$$\exp\left(-\alpha\left(\frac{(\theta-\tau)}{(1-\beta\tau)}\right) + (1-\beta)\delta(\theta-\tau)\right) \quad \theta \geq \tau$$

$$E(\theta) = 0 \qquad \theta \leq \tau \tag{3}$$

Where α is the number of stirred tanks, β the fraction of flow which enters the gamma mixing section, τ the dimensionless residence time of the plug flow reactor. Again, the resulting RTD function follows a mono-modal curve, thus not being able to describe and predict internal recirculation.

The Markov-chains discrete time model considers a random behavior of fluid elements [25]. In this model, ideal CSTR and plug flow reactors are connected to each other in different arrangements but based on a specific algorithm [25-27]. Theoretically the Markov-chain model may result in multimodal RTD curves. However, this aspect of the model has been ignored till now and is not the subject of the present study. It is noted that the model presented by Sohrabi and Marvast was not able to predict the experimental data created in the present work for a complex impinging streams reactor system [25].

The Direct Simulation Monte Carlo Method (DSMC method) was applied by *Sohrabi et. al.* in 2008 to formulate the dynamic behavior of fluid in a simple impinging streams reactor system. There, the RTD was determined based on the Boltzmann equation using a direct Monte Carlo simulation method. It was reported that DSMC model is more accurate and flexible than the Markov-chains discrete time model which had been already applied by *Sohrabi et al.* [25-27] and the agreement degree between experimental data and those predicted by DSMC model was within 85%, while in case of the Markov-chains discrete time model the degree of agreement was 75% [22]. However, the DSMC method is unable to create multi-modal RTD curves.

New theoretical model

Here we propose a general model that is able to simulate impinging reactors with a notable flexibility. The main component of the model is a series of an ideal PFR and a recycle reactor. The recycle reactor itself consists of a finite number of identical ideal CSTR's (number = p) each with an equal space time τ_{CSTR} and a recycle stream of a recycle ratio equal to R. Fig. 3 illustrates this main component which is denoted as "main row".

The raison d' etre of the PFR unit is rather rational as the real reactor geometry is cylindrical. However, the impinging streams may create recirculation currents within the system and the CSTR's in series plus the recycle stream take this fact into account. The ideal PFR acts as a 'distributor' and the CSTR's zone (with recycle stream) acts as recirculation generator.

The general model is simply a replication of the main component (main rows) in a parallel fashion. The main feed volumetric flow rate will be distributed through the parallel streams by fractions denominated as f_i, where i is the number of the parallel stream. The overall scheme is shown in Fig 4.

The parameters of the proposed model are as the following:

$\tau_{i,PFR}$: Space time of ideal PFR in row i

$\tau_{i,CSTR}$: Space time of ideal CSTR in row i

p_i: Number of ideal CSTR's in row i

f_i: Fraction of feed flow rate entering row i

The replication of the main row shown in Fig. 3 allows for the appearance of multiple recycling zones within the reactor volume, actually in accordance to the experimental evidence discussed in the following sections.

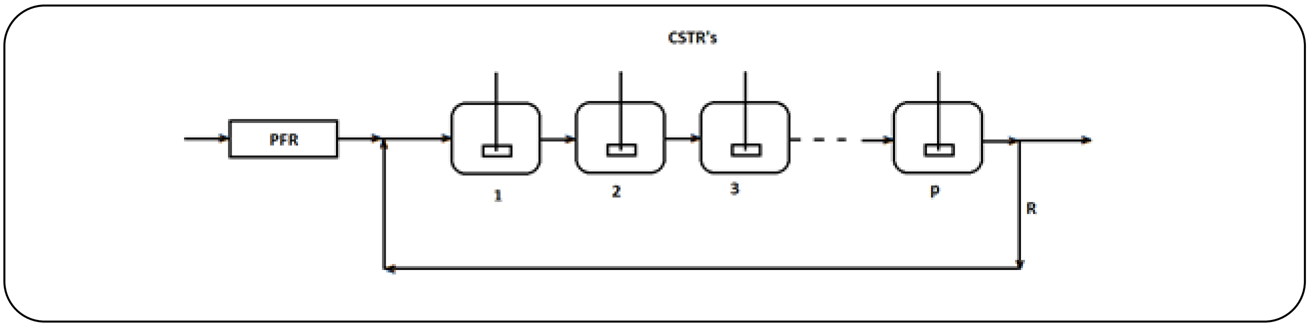

Fig. 3. Schematic illustration of the main row.

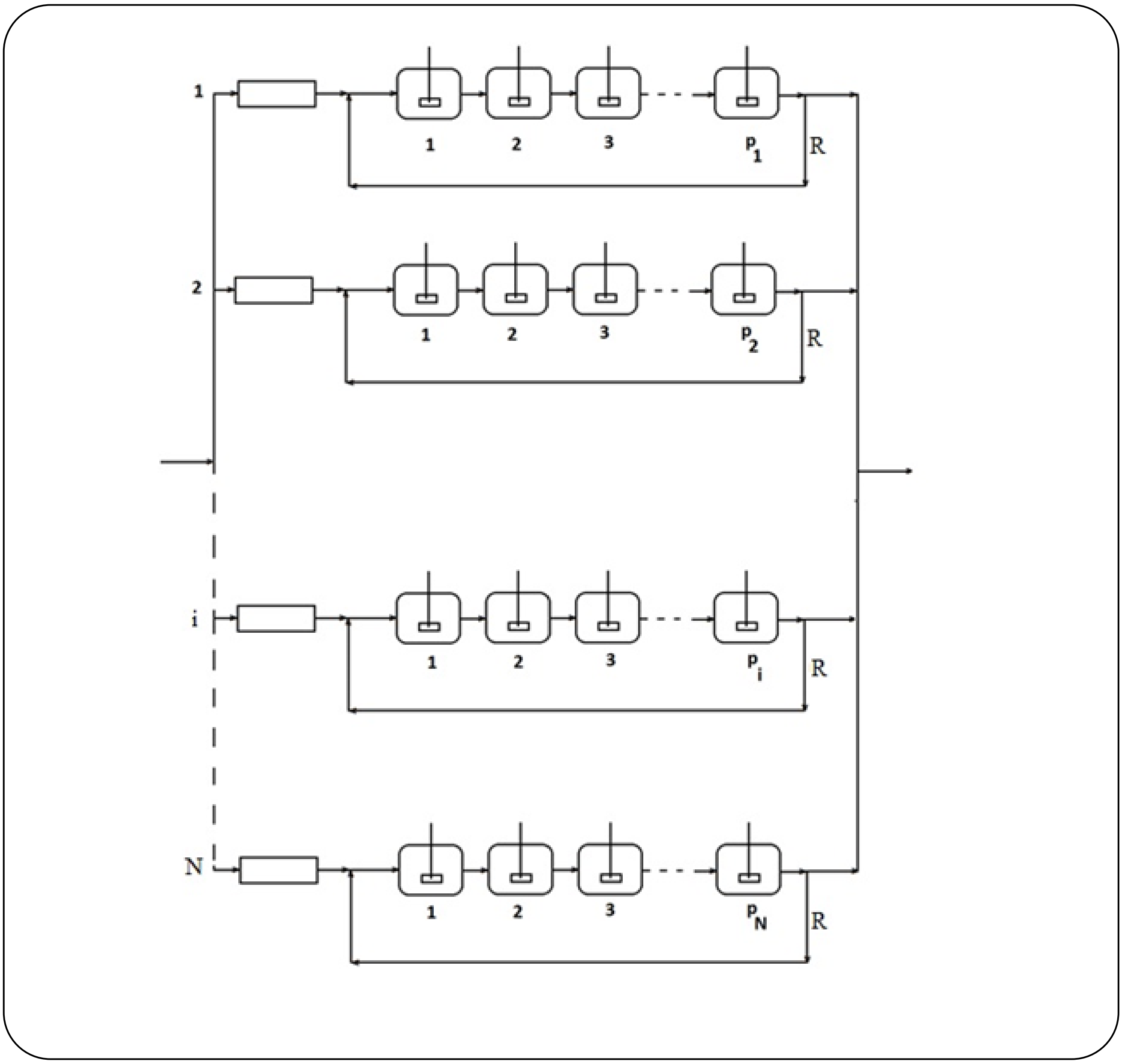

Fig. 4: Proposed overall scheme of the reactor system.

At this stage, we reformulate the model in the "s" domain, by considering the Laplace transform of the governing equations. Fig. 5 demonstrates such visualization.

where:

$$G_{i,0}(s) = e^{-\tau_{i,PFR},s} \quad (4)$$

$$G_{i,T}(s) = \frac{1}{R+1}\sum_{n=0}^{\infty}\left(\frac{R}{R+1}\right)^n\left(\frac{1}{1+\tau_{CSTR}s}\right)^{p_i(n+1)} \quad (5)$$

The inverse Laplace transform of Equation (5) is as follows:

$$E_{i,T}(t) = \quad (6)$$

$$\frac{1}{R+1}\sum_{n=0}^{\infty}\left(\frac{R}{R+1}\right)^n \frac{\tau^{n\cdot p_i+p_i-1}}{\tau_{CSTR}p_i(n+1)}\frac{e^{-\frac{\tau}{t_{CSTR}}}}{(n\cdot p_i+p_i-1)!}$$

For each row, the resulting E_i (t) is simply:

$$E_i = \delta(t-\tau_{i,PFR})\cdot \quad (7)$$

$$\frac{1}{R+1}\sum_{n=0}^{\infty}\left(\frac{R}{R+1}\right)^n \frac{\tau^{n.p_i+p_i-1}}{\tau_{CSTR}p_i^{(n+1)}}\frac{e^{-\frac{\tau}{\tau_{CSTR}}}}{(n.p_i+p_i-1)}$$

The overall exit time distribution function is then:

$$E(t) = \sum_{i=1}^{N} f_i \cdot E_i(t) \quad (8)$$

In order to effectively reduce the large number of controlling parameters present in the model, the following assumptions have been made:

1- The number of CSTR's and their corresponding recycle ratio is the same for all branches.

2- The residence time of each of the CSTR tanks in all rows is the same for all branches and equal to τ_{CSTR}.

3- The number of branches, N, is not a parameter. It may selected equal to the maximum number of peaks (recycling peaks) observed throughout the different experiments run. In our case this is equal to 5. However, N might be taken any large number and this makes no changes in the results due to the equation used for distributing the flow between the branches (see below, case 4). Actually, we have observed that in any case of total volumetric flow implemented, the "recirculating" peaks turn out not to appear at constant intervals or even, to appear again at longer times. Imposing new branches, imposes a new peaks.

The method of guessing the number of branches may also be considered a mathematical trick to ascertain a flexible solution to the problem.

4- The fraction of volumetric flow entering each branch is calculated by the following relation:

$$f_i = \exp(-ki) \qquad i \geq 2 \quad (9)$$

$$f_1 = 1 - \sum_{i=2}^{N}\exp(-ki) \quad (10)$$

where k is a constant parameter.

Preliminary calculations based on nonlinear regression without implementing any formula for the calculation of fi showed that this parameter decreases sharply as i increases. Such a dependency on i may be thus considered, as a first attempt, as an exponential decrease. Equation 9 has been adopted according to this explanation. . Equation 10 is a trivial consequence of the fact that the sum of all f_i's should be one.

5- The residence time of the PFR in row 1 is zero. The value of this parameter changes with the number of the rows by the following relation embracing the periodicity of the recycling phenomenon, i.e.:

$$\tau_{i,PFR} = (i-1)\tau_{PFR} \qquad i \geq 2 \quad (4)$$

The idea behind the proposed criterion lies in the fact that such an equation imposes the creation of the multiple peaks observed in the RTD even in the case of absence of recirculation.

According to this assumptions, the number of unknown parameters is limited to 5 ones, namely k, τ_{CSTR}, τ_{PFR}, p and R. It should be reminded that the initial guess for the value of τ_{PFR} may be obtained finding the time in the RTD which the first recirculation peak is observed.

RESULTS AND DISSCUSSION

Fig. 6 shows the RTD curve of the 3-nozzle impinging streams reactor system subject of the present study as a function of total flow rate. It is observed that the RTD curve depend strongly on the flow rate. On the other hand, the curves are generally multimodal, with a rather complex asymmetric shape. The existence

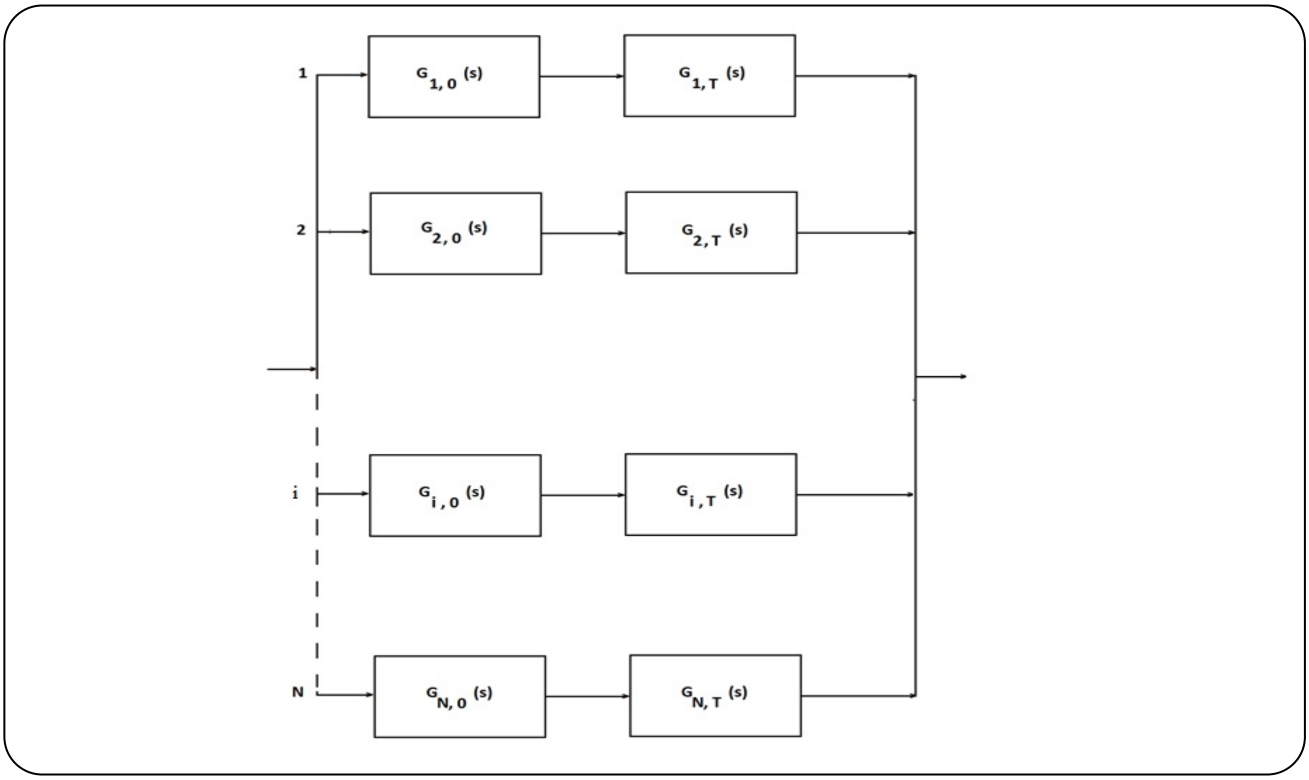

Fig. 5: The overall proposed scheme shown in the "s" domain.

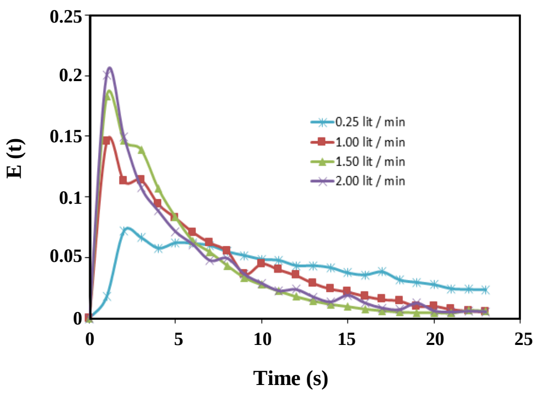

Fig. 6: Experimental residence time distribution of the impinging stream reactor, E(t).

of several peaks most probably testifies the existence of recirculation streams within the reactor volume.

The depicted RTD curves are not predictable by the conventional models such as tank in series, GDB, DSMC and Markov-chains discrete time models applied so far for simulating simpler impinging streams reactor systems. The complex oscillations found in the tail of the experimental curves of this study could not be predicted by these models.

Based on Equations (7) and (8), a code in MATLAB was written that predicts the overall exit residence time distribution function of the systems under investigation. The controlling parameters k, τ_{CSTR}, $\tau_{1,PFR}$, p and R have been optimized in order to fit the experimental data of the exit residence time distribution function.

Figs. 7 to 10 show the model and experimental data for each flow rate of injection. The agreement between model and experimental data is reasonable.

Fig. 11 shows the fraction of the total inlet stream entering each branch as a function of volumetric flow rate. It turns out that the main portion of the total inlet stream (more than 86 % in all cases) enters row 1 and the other routs consist a small portion of the total liquid flow rate fed to the system.

It is observed that increasing the total inlet flow rate results in a mild decrease of f_1 and corollary increase of fractions f_1 to f_2. In other words, the complexity of the impinging streams reactor system is lowest at highest total inlet streams flow rate. As the flow rate is decreased, it looks as if the liquid volume within the reactor tends to segregate (although mildly) and create non-interacting zones that behave separately and exit the system individually.

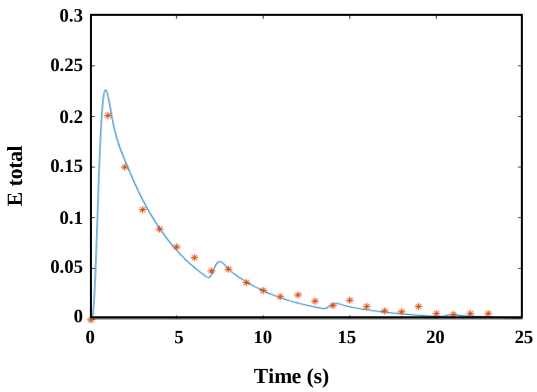

Fig. 7: Model and experimental data for the RTD at a total flow rate of 2.00 L/min.

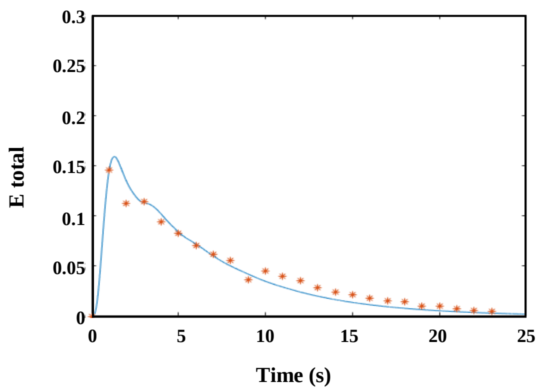

Fig. 9: Model and experimental data for the RTD at a total flow rate of 1.00 L/min.

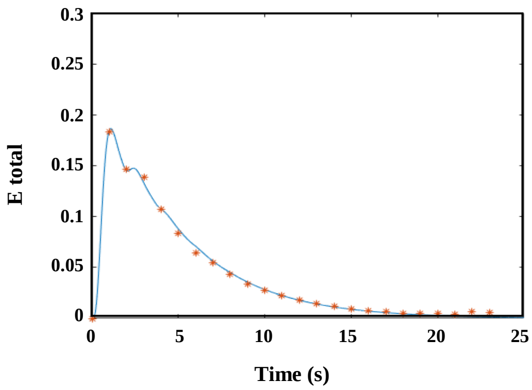

Fig. 8: Model and experimental data for the RTD at a total flow rate of 1.50 L/min.

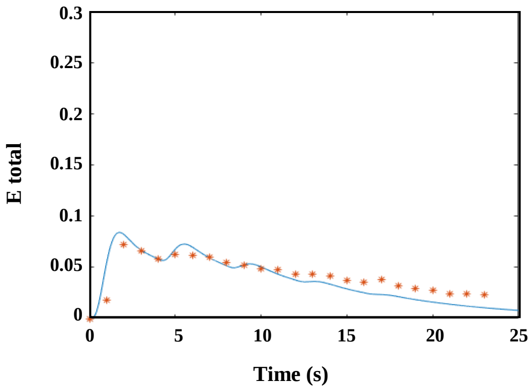

Fig. 10: Model and experimental data for the RTD at a total flow rate of 0.25 L/min.

It is well known that the hydrodynamic state of a fluid within a reactor system lies between two well defined boundaries of 'maximum mixedness' and 'completely segregated' [36]. Our verified model shows that for the three nozzle impinging streams reactor system subject of the present study, the degree of fluid segregation within the system is a weak function of total the flow rate.

Table 1 shows the optimum parameters obtained for the controlling parameters as a function of flow rate.

It may be claimed that the results of the new mathematical model are consistent with the physics of the system and this is explained in what follows. Considering the values obtained for the different parameters of the system for the different total flow rates understudy, a schematic two dimensional (2D) representation of the possible hydrodynamic state within the reactor system has been depicted (Fig. 12). Zone 1 corresponds to a major portion of the total area and is presumed being created mainly by the axial inlet stream which constitutes half of the entering total flow rate. The axial inlet stream undergoes the minimal perturbation due to the tangential streams. However, the tangential streams add partially to this stream, making zone 1 consisting more than 86 % of the total zones. Areas 2, 3, 4 and 5 represent a minor part of the total flow within the reactor system. They are closer to the inner cylindrical wall of the reactor. It is presumed that the tangential streams add mostly to the axial inlet stream, leaving low volume areas 2 to 5 behind. The impingement of the tangential streams with themselves and the axial stream is highly desirable and takes place in the flow rate range of 1.0 to 2.5 L/min, based on the large value of p and R calculated through mathematical modelling. This has been intentionally shown as rotating arrows within the shaded areas.

Table 1: Optimum parameters for the controlling parameters as a function of flow rate. SSE is the sum of squared errors between the predicted and experimental points.

flow rate (lit / min)	*k*	τ_{CSTR}	$\tau_{1,PFR}$	p	R	SSE
0.25	.8	0.40	4.0	5	3	0.0095
1.00	1.4	0.30	2.5	5	3	0.0226
1.50	1.3	0.25	1.8	5	3	0.0374
2.00	1.2	0.20	6.7	5	3	0.0540

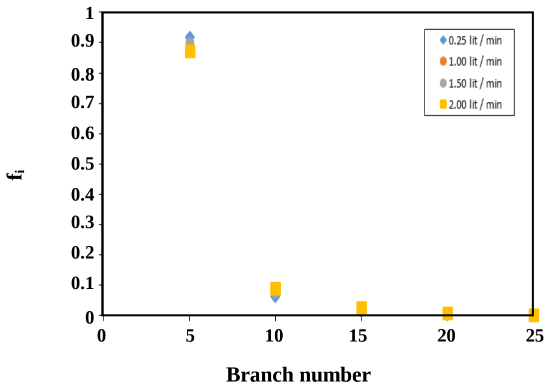

Fig. 11: Fraction of inlet stream entering different branches as a function of total flow rate.

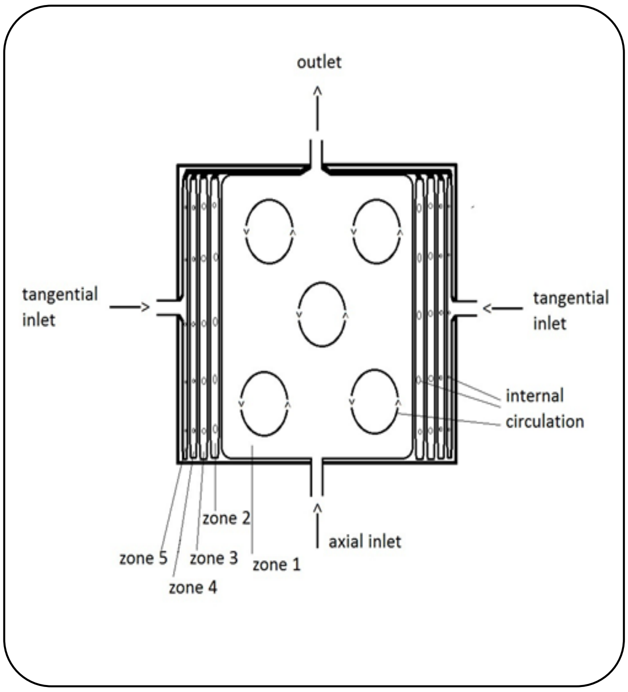

Fig. 12: Two dimensional schematic representation of the possible hydrodynamic state within the impinging streams reactor system consisting of one axial and two tangential inlet streams.

CONCLUSIONS

In the present work we have proposed for the first time a relatively simple model for modeling impinging streams reactors based on simple theoretical criteria. The model allows for time delays due to ideal PFR reactors, and at the same time, allows for peak broadening due to the existence of CSTR's in series. More importantly, a recirculation implemented on the CSTR's ensures the prediction of recirculation peaks. To make the model more flexible, such an arrangement is allowed to be repeated many times, although with an effect on the total RTD decreasing exponentially. Using Laplace Transform, the RTD function of the proposed complex system of ideal reactors for the three inlet nozzles impinging streams reactor understudy turns into an algebraic relation allowing high precision computation avoiding any integration or derivatization operation. The model has only 5 controlling parameters that allows it to simulate complicated RTD curves.

Summing up, a new mathematical model has been developed that is able to tackle recirculating streams within the reactor volume.

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