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Hydraulic Regime Study for a Bench-Scale Electrochemical Arsenic Remediation (ECAR) Reactor Utilizing Methylene Blue as Tracer

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Abstract- *Long-term exposure to low level arsenic is becoming a very serious health problem now-a-days. Electrochemical Arsenic Remediation, (ECAR) is a low-cost, low-maintenance and robust technology that efficiently removes arsenic from the groundwater. Considering the targeted demand of providing arsenic-safe water, to a large section of arsenic-affected population as the community-level drinking water, the development of continuous-scale ECAR unit has been in progress. Initially a laboratory scale (35L) continuous flow type ECAR reactor has been fabricated whose hydraulic characteristics is being investigated in the present study with methylene blue dye as the tracer. This reactor has been characterized using the residence time distribution (RTD) data that allows identifying the flow pattern and its deviations. The cumulative distribution function curves are being plotted with the help of RTD data, which exhibit the behavior of the reactor similar to that of a non-ideal CSTR.*

Key words- *Electrochemical arsenic remediation (ECAR), continuous flow, pulse input, tracer study, residence time distribution, mean residence time, continuous stirred tank reactor*

I. INTRODUCTION

Arsenic in drinking water is a major public health problem threatening the lives of over 140 million people worldwide [1, 2]. The acute toxicity of arsenic in high concentrations had been known for centuries, but only recently a strong adverse effect on health has been reported which is associated with long-term exposure to a very low level arsenic [3]. The range of arsenic concentrations found in natural waters is large, varying from less than 0.5μg/L to more than 5000μg/L [4]. Increased knowledge of the cancer risk led the World Health Organization (WHO) to lower its recommended maximum limit of arsenic from 50μg/L to 10μg/L in drinking water [5]. An interdisciplinary research conducted by Gadgil Lab, University of California, Berkeley has led to the development of an efficient, effective, and electricity-based, emergent technology known as Electro Chemical Arsenic Remediation (ECAR) [6], which is a promising technology that is compatible with a community clean water center implementation model and has the potential to operate at extremely low cost. ECAR has been shown to work in 3L as well as 20, 100 and 600L batch prototypes so far. In order to run technical trials in field, a prototype is needed that is capable of treating a larger volume of water. Field trials are already done with the 100 and 600L prototypes in batch mode. Considering the targeted demand of providing 10L of arsenic-safe water, per person per day to a large section of arsenic-affected population as the community-level drinking water, the development of continuous-scale ECAR unit has been in progress. At the very initial stage of this part of research, a laboratory scale continuous type ECAR reactor has been fabricated whose hydraulic characteristics is to be investigated as an important prerequisite.

Hydraulic regime study has been useful in gaining information regarding the internal flow paths within a treatment reactor system. The presence of preferential flow paths and dead zones can be identified through the introduction of a tracer at the inlet followed by spatial monitoring of tracer concentrations throughout the reactor. Spatial tracer monitoring can be conducted along longitudinal, lateral and vertical profiles within a reactor to gain an insight into the distribution of flow velocities, preferential flow paths and mixing characteristics as water moves through the system [7, 8, 9, 10, 11, and 12].

In hydraulic analysis, any substance can be essentially used as a tracer, provided that it is highly soluble in water (representative), does not react with the water and any of its constituents (preferably inert and conservative in nature) occurs in low background levels within the reactor, is relatively easy and inexpensive to analyze, has low toxicity, and does not influence the flow pattern in a significant way [13, 14]. One of the most important tests of a tracer's reliability is the recovery percentage. Consequently, the tracer mass recovery rate should always be reported in the results of a tracer study. It is generally considered acceptable if at least 80% of the mass of tracer added as an impulse at the inlet is recovered at the outlet.

Dyes have advantages of low detection limits, zero natural background and low relative cost. However, they are susceptible to a variety of environmental influences which can affect their stability and detection [15, 16]. A number of studies of dye-stained preferential pathways have been published in the literature [17, 18, 19, 20, 21, and 22]. Under the purview of the present study, a 35 L prototype of continuous flow ECAR reactor has been fabricated which will be analyzed with electrochemical system models to permit the prediction of performance of a full-scale device. The study of the hydraulic behavior of this reactor is of fundamental importance for analyzing the efficiency of any full-scale arsenic remediation system. The hydraulic characteristics of this bench-scale, continuous-stirred tank reactor was studied using some major operating parameters such as, liquid flow rate, time-concentration data, residence time distribution function *E(t)*, the mean residence time *(t_m)* and cumulative distribution function *F(t)*. In the present study, we attempted to characterize this reactor using the residence time distribution data. The determination of experimental residence time distribution (RTD) is a very effective technique that allows identifying flow pattern deviations [23].

II. MOMENTS OF THE STUDY

In any reactor, the time the atoms have spent in it is called the ***residence time*** of the atoms in that particular reactor. The distribution of residence times can significantly affect the performance of the reactor irrespective of its type. The ***residence-time distribution*** (RTD) of a reactor is a characteristic of the mixing that occurs within the chemical reactor. The theory of residence time distributions generally begins with three assumptions:

1. The reactor is at steady-state
2. Transports at the inlet and the outlet takes place only by advection, and
3. The fluid is incompressible

The CSTR is thoroughly mixed (both axially and horizontally) and possesses a far different kind of RTD than the plug-flow reactor. The residence time distribution function *E(t)* is expressed as:

$$E(t) = \frac{C(t)}{\int_0^\infty C(t)dt}$$

Here, *C (t)* is the concentration at the time t.
The integral in the denominator is the area under the *C* curve. An alternative way of interpreting the residence-time function is in it integral form:

$$\begin{bmatrix} fraction\ of\ material\ leaving\ the\ reactor \\ that\ has\ resided\ in\ the\ reactor\ for\ the \\ time\ between\ t_1\ and\ t_2 \end{bmatrix} = \int_{t_1}^{t_2} E(t)\,dt$$

We know that the fraction of all the material that has resided for a time *t* in the reactor between *t* =0 and *t* =∞ is 1;
Therefore,

$$\int_0^t E(t)dt = 1$$

The fraction of the exit stream that has resided in the reactor for a period of time shorter than a given value *t* is equal to the sum over all times less than *t* of $E(t)\,\Delta t$, or expressed continuously,

$$F(t) = \int_0^t E(t)dt$$

This equation is defined as ***cumulative distribution function*** *and called it F(t)*. We can calculate *F*(*t*) at various times *t* from the area under the curve of an *E*(*t*) versus *t* plot.

III. MATERIALS AND METHODS

A. Experimental design

Reactor Configuration: The single stage reactor, used for the present experimentation programme, was fabricated from a fiber glass sheet 0.635 cm. thick. The final configuration of the reactor is shown in the following *Figure 1, 2, 3*and 4.

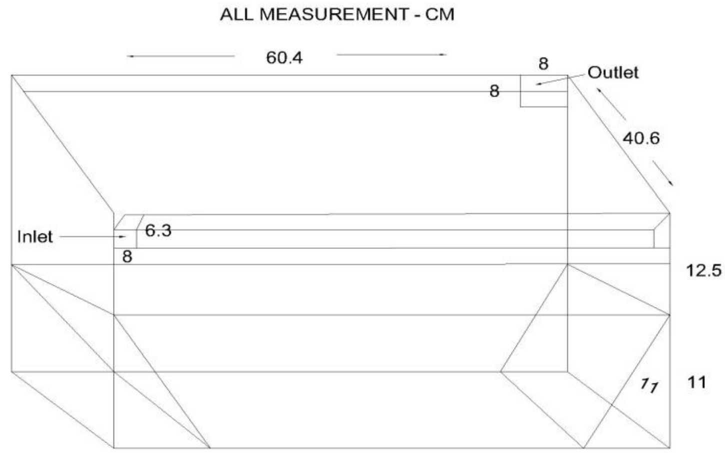

Figure 1. Front view of lab scale ECAR reactor

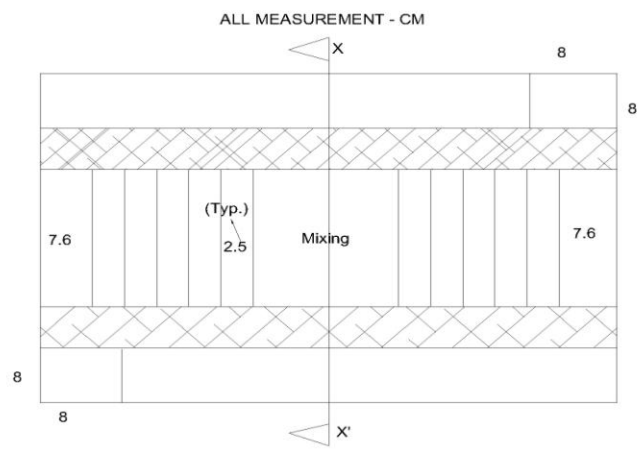

Figure 2. Top view of lab scale ECAR reactor

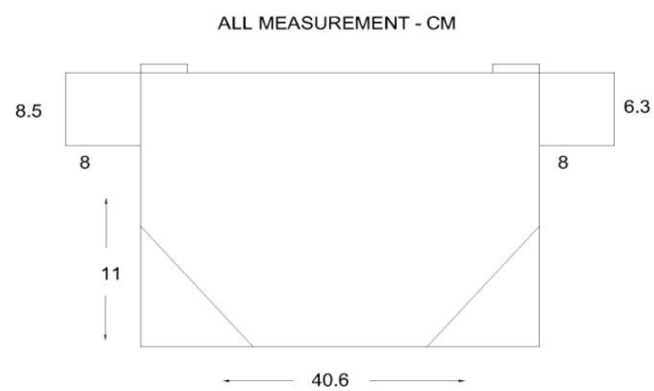

Figure 3. Cross sectional view of lab scale ECAR reactor

Figure 4. Experimental Lab scale ECAR reactor

The trough of the reactor was 60.4 cm. in total length, 40.6 cm in width and 23.5 cm in depth with a trapezoidal cross-section at the lower part. The reactor has the liquid holding capacity in the range of 25- 40 L. At the middle of the reactor there is a propeller with varying rotational speed and in the both sides of the propeller there are 6 fiber glass plates, each having a length of 21.8 cm, breadth of 24.3 cm, and thickness of 0.15 cm. All the fiber glass plates are closely fixed at the bottom with two wooden pieces at a spacing of 2.54 cm so that the plates do not swing with the turbulences created during the flow. The outlet arrangement was made by fitting a 0.59 cm diameter pipe, adjusted to a particular depth having a particular degree of submergence. Cleaning of the reactor was done as per requirement with cleansing solution only. In the study, the inflow and outflow rate were maintained constant for a particular set of experiment.

Dye solution: In the present study, methylene blue has been used as a tracer for the convenience of its sampling and testing in the laboratory. Methylene blue is a heterocyclic aromatic chemical compound with the molecular formula $C_{16}H_{18}N_3SCl$. At room temperature it appears as a solid, odorless, dark green powder that yields a blue solution when dissolved in water. It is highly soluble in water and can be easily detected. The concentration of the dye has been maintained at a constant 5ppm level throughout the experimental study. λ_{max} value for methylene blue was found to be 664 nm from the absorption spectrum data [24].

B. Tracer experiment procedure:

In each and every run, the desired flow of the liquid into the reactor was fed by pulse-injection method i.e. a fixed amount of tracer was suddenly injected in one shot into the feed stream entering the reactor in as short time as possible. The samples were collected at the outlet at different intervals and tested for dye concentration as per standard methods with Varian UV-visible spectrophotometer (Model no: Cary 50 Bio: Serial no: EL06013314). The operation was continued until the dye concentration at the outlet end reached its initial concentration.
The extent to which non-ideal flow exists in a given reactor is indicated by the RTD. However, because of the difficulty in knowing how long individual molecules of water have spent in a reactor, the RTD is typically inferred by studying the behavior of a soluble, inert tracer on passage through the reactor. The tracer is assumed to follow the same flow pattern as the parcel of water with which it entered the reactor, and should therefore give a reasonable reflection of the hydraulic RTD. The resultant tracer RTD can then be used to elucidate the actual reactor water volume that is involved in treatment (hydraulic efficiency), as well as the degree of apparent mixing and deviation from ideal flow. The residence time distribution function and its moments are calculated from the time-concentration or *C* curve.

IV. RESULT AND DISCUSSION

In the present study, we have obtained different t_m (the mean residence time) for different flow rate and mixing pattern and in each case we have got specific result, which can be explained with adequate logical support. For instance, if t_m is large, there will be a slow decay of the output transient, $C(t)$, and $E(t)$ for a pulse input. On the contrary, if t_m is small, there will be rapid decay of the transient, $C(t)$, and $E(t)$ for a pulse input. In our study, we have been often exposed to such situations. When the mean residence time i.e. t_m is small, the concentration of the tracer dye was decreasing at a rate higher than that occurring at high t_m value. In addition to this, the RTD can also be used to predict conversion in existing reactors when a new set of reaction being tried in an old reactor. With the help of these RTD data we can also explore the arsenic removal capacity of ECAR in the ground water environment under continuous mode of operation. However, RTD is not unique for a given system, and we need to develop models for the RTD to predict the conversion. The following data are presented in the tabular form for the different sets of experiments:

Table 1. Tracer data with different flow rate and propeller speed keeping other operational parameters constant (with no recirculation, only propeller is used for mixing) *(Dye concentration = 5ppm; pH =7.0; Number of plates = 12 with 6 plates on each side)*

Exp No.	Total time	Flow rate (L/min)	Maximum absorbance at the time (min)	Area (min.mg/L) from C(t)/t curve	Area from E(t)/t curve	t_m
1	90	0.67	5	196.863	1	39.49
2	90	0.75	9	157.913	0.999	35.989
3	90	0.57	6	73.935	0.999	34.784
4	90	0.6	6	127.375	0.944	31.119
5	90	0.6	7	175.717	0.999	37.330
6	90	0.7	7	145.794	0.999	34.589
7	90	0.87	6	155.266	0.999	35.646
8	90	0.87	6	45.534	1	20.412
9	90	0.81	9	150.575	1	34.538
10	90	0.81	11	132.864	1.012	31.238
11	90	0.77	10	140.800	1.001	34.968
12	90	0.52	13	147.729	1	41.277
13	90	0.57	14	190.617	1	43.179
14	90	0.57	8	130.504	1	31.481
15	90	0.47	21	211.658	1	41.912
16	90	0.45	9	167.426	1	40.567
17	90	0.45	12	177.758	1	40.752
18	90	0.49	11	205.0173	1	40.764
19	90	0.46	10	189.42	1	39.998
20	90	0.46	15	196.834	1	41.776
21	270	0.4	17	358.85	1	89.832
22	90	0.4	17	208.508	1	37.96
23	90	0.6	10	177.986	1	38.918
24	270	0.55	13	275.358	1	59.443
25	120	1	6	140.806	1	30.452
26	90	0.83	8	157.872	1	33.167
27	150	0.74	10	191.814	0.999	40.757
28	90	0.8	8	155.803	1	34.366
29	90	0.95	5	143.878	1	31.412
30	90	0.93	8	157.620	0.936	33.384

The cumulative distribution function i.e. F(t) is shown graphically in the following figure:

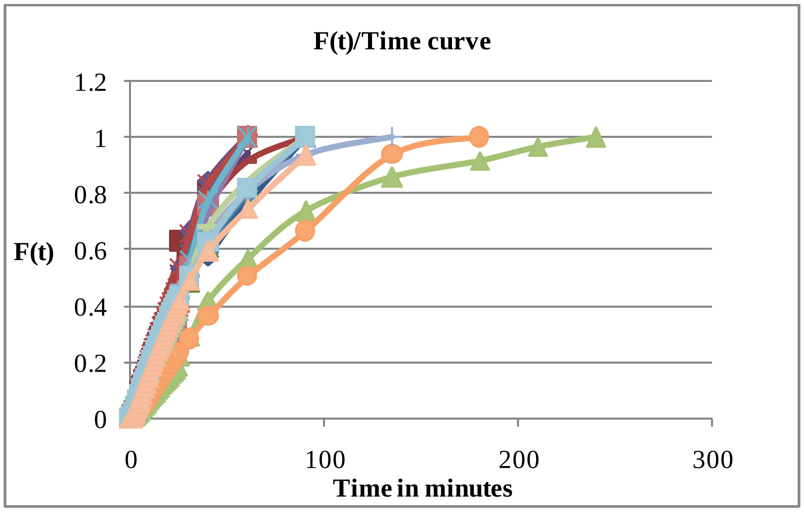

Fig 5. Cumulative distribution curve (when propeller was used for mixing) representing Table 1.

In the first thirty sets of experiments we have used a propeller for mixing and got the above type of graph. However, in the subsequent sets of experiments we have used a submersible pump for recycling the water in the middle of the reactor for mixing and obtained the following characteristics of the data:

Table 2. Tracer data with different flow rate and propeller speed keeping other operational parameters constant (recirculation is done with a submersible pump) *(Dye concentration = 5ppm; pH =7.0; Number of plates = 12 with 6 plates on each side)*

Exp No.	Total time (min)	Flow rate (L/min)	Maximum absorbance at the time (min)	Area (min.mg/L) from C(t)/t curve	Area from E(t)/t curve	t_m (min)
1	60	0.9	5	79.875	0.99968	23.034
2	120	0.7	6	181.743	1	38.422
3	120	0.95	7	128.326	1	26.155
4	120	0.58	11	206.572	1	46.313
5	120	0.66	14	147.221	1	31.519
6	120	0.75	8	219.747	1	39.253
7	120	0.5	9	202.0594	1	43.609
8	120	0.4	20	271.985	1	54.127
9	120	0.45	14	239.578	1	50.66
10	120	0.51	12	246.688	1	54.103

The cumulative distribution function i.e. F(t) is shown graphically in the following figure:

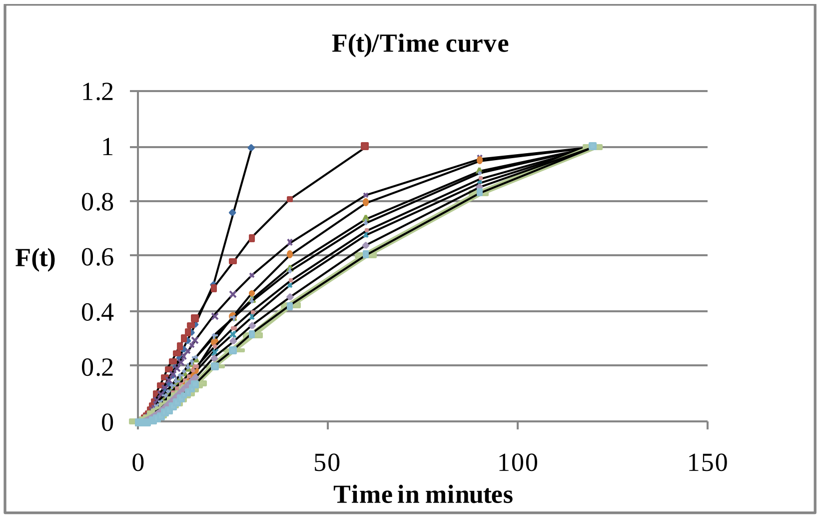

Fig 6. Cumulative distribution curve (recirculation was done with pump) representing Table 2.

In the next set of experiments we have run the same set of experiments in continuous mode but we have collected the samples from different parts of the reactor at the same time with same intervals. The positions of the sampling points are mentioned in the table 3 below.

Table 3. Details of Sampling Locations

Sample Number	**Samples taken from the distance measured from the left face of the reactor (cm)**
1	**7.62**
2	**12.7**
3	**30.23**
4	**47.76**
5	**52.84**
6	**outlet**

At this phase of study, the inflow rate of the tracer was varied as per the table 4 furnished below.

Table 4. Details of inflow rate of the tracer

Set No.	1	2	3	4	5	6	7	8	9	10
Inflow Rate of tracer (lpm)	0.89	0.9	0.63	0.44	0.79	0.7	0.6	0.7	0.9	0.8

The results obtained for this phase of study are furnished in the following plots.

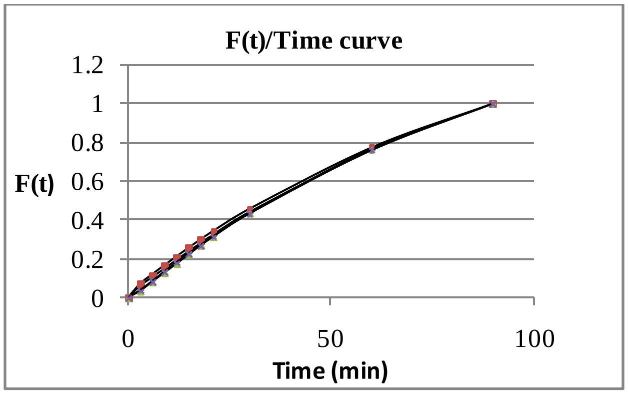

Fig 7. Cumulative distribution curve for set-1

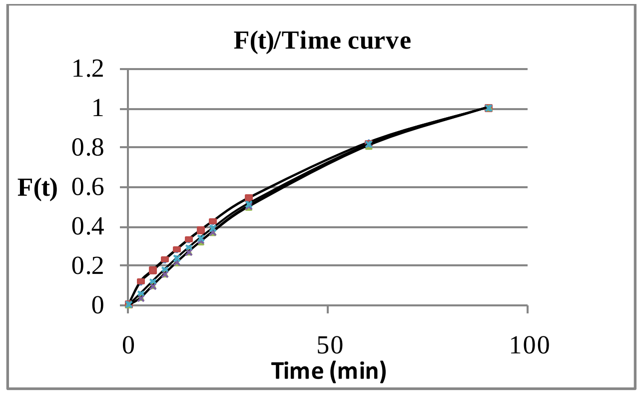

Fig 8. Cumulative distribution curve for set-2

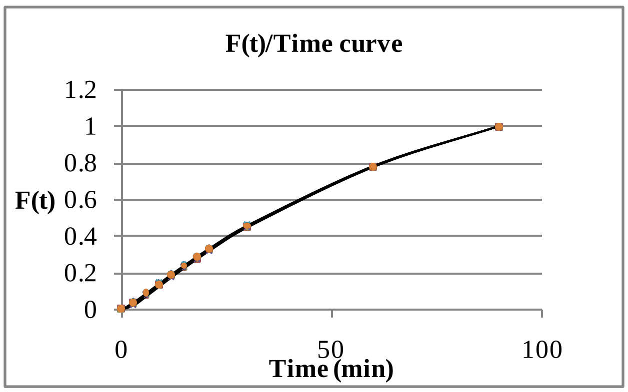

Fig 9. Cumulative distribution curve for set-3

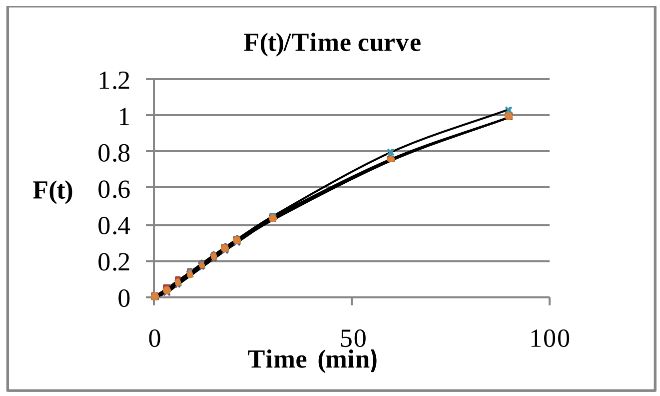

Fig 10. Cumulative distribution curve for set-4

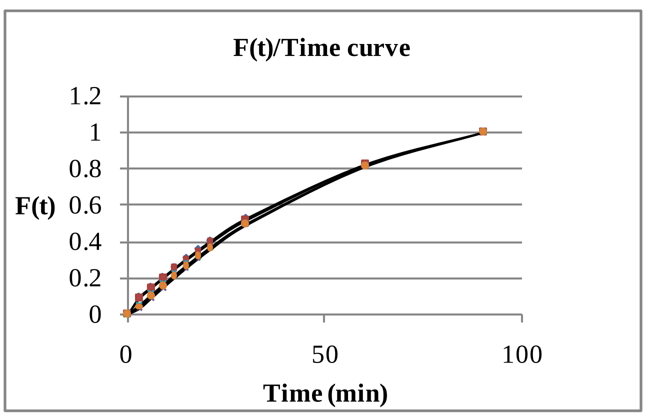

Fig 11. Cumulative distribution curve for set-5

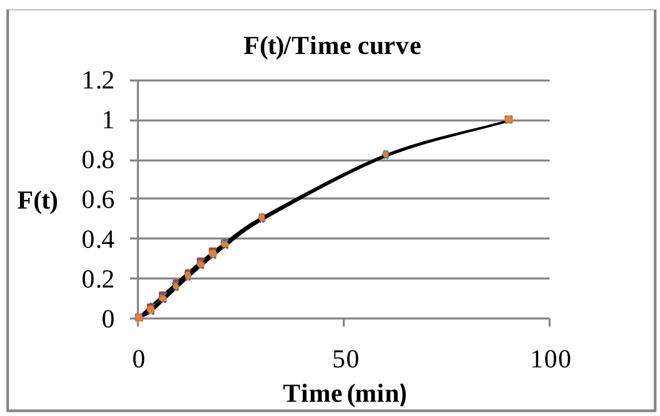

Fig 12. Cumulative distribution curve for set-6

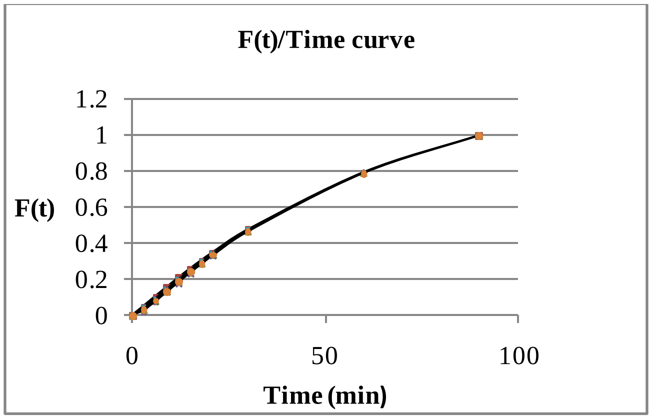

Fig 13. Cumulative distribution function for set-7

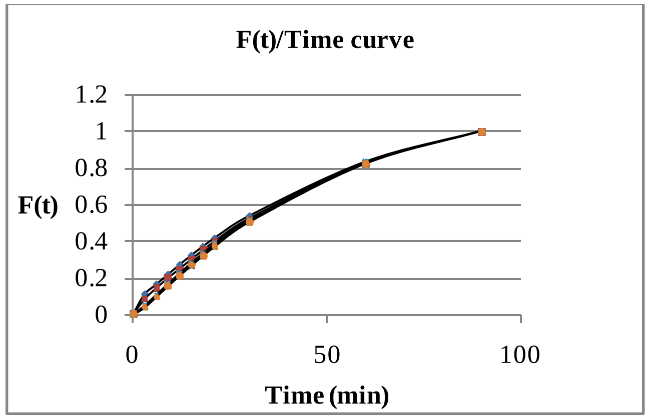

Fig 14. Cumulative distribution function for set-8

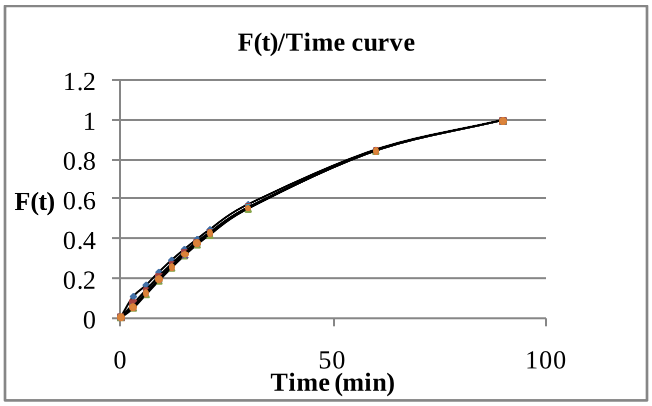

Fig 15. Cumulative distribution function for set-9

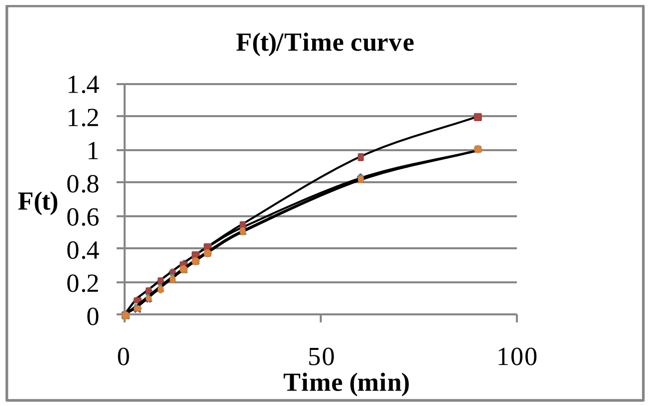

Fig 16. Cumulative distribution function for set-10

Though the samples were withdrawn from different locations in the reactor, the nature of the curves exhibits flow pattern of almost identical characteristics. It is evident from these graphs that, in the present experimental reactor system, there is a highly agitated zone in the vicinity of the impeller that can be modeled as a nearly perfectly mixed CSTR system.

V. CONCLUSION

The experimental work was carried out successfully to define the hydraulic regime of a continuous reactor. This reactor exhibits a non-ideal CSTR behavior. Based on the hydrodynamic studies a prediction model can be developed. The experimental data showed a good fit to the theoretical model and verified the utility of this model to scale-up the process. It is noticeable that the continuous stirred tank reactor (35L) is able to operate without operational problems with no dead volume, bypassing and turbulence. It points out the relevance as well as the novelty of the results obtained in the present work.

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