Industrial Scale RTD Measurement Using Gold Radiotracer

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ABSTRACT: *Residence Time Distribution (RTD) is a suitable method to find out the hydrodynamics of any industrial or lab-scale reactor. Radiotracer ^{198}Au was used to trace the liquid phase of the industrial scale continuous three tube pulp digester. The radiotracer was injected instantaneously as an impulse input in the liquid phase and concentration versus time data were collected at the inlet and outlet of each tube using a data acquisition system. The flow modeling of continuous pulp digester has been done in two ways. Firstly, the whole digester was considered a single reactor. Secondly, each tube of the digester was considered a single reactor separately. For the second case, the convolution procedure was opted to deal with a non-ideal input signal for the second and third tubes. Axial dispersion model and tank in series with a back-mixing model were used to simulate the experimental data. Mean residence time and Peclet numbers were calculated for each tube and the whole digester.*

KEYWORDS: *Au-198, Axial dispersion model; Convolution; Pulp digester; Radiotracer; Tank in series with a back-mixing model.*

INTRODUCTION

The paper industries are considered as an ever-growing sector in the world including developed as well as in developing countries. The world's total production of paper and other paper products is more than 400 million tons out of which, India produced 15 million tons [1]. Nearly 24% of the paper products are made from wood. Only 11 % of the paper is produced from non-conventional raw materials such as wheat straw, rice straw, bagasse, etc. [2]. In India, the demand for paper and other paper products is increasing continuously, conversely ever decreasing forest area is hitting badly to the growth of the paper industry. Hence, importance is given to the fast-growing and easily available non-conventional raw materials like bagasse, wheat straw, rice straw, etc. These bio-cellulosic raw materials are easily available as agro-residue or byproducts of farming [2,3].

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Furthermore, the continuous pulping process is more favorable to achieve less variation in pulp quality as compared to the traditional batch process. For a continuous process, Continuous tube digesters are more appropriate to carry out the chemical pulping using the different types of raw material. Each tube of the continuous digester is pressurized and this high pressure and temperature are maintained through injecting steam. A detailed explanation is about the pulping process using a continuous digester is already available in the literature [4, 5].

The basic aim of the pulping process is to remove maximum lignin from the cellulose-rich raw material without affecting the cellulose fiber's strength and to maintain less variation in kappa number. Kappa number is the measure of an amount of delignification. The efficiency of the pulping process is based on the operating parameters and the time spent by the process material inside the digester. Residence time distribution can provide vital information about hydrodynamics, malfunctioning, or a fault inside the digester. The residence time of the digester can be controlled by controlling the speed of the conveyor fitted inside the tubes of the digester.

Different types of malfunctioning like bypassing, channeling, dead volumes, etc. can be determined using RTD study. This technique is widely used for the identification of the above-mentioned malfunctioning and measurements of various parameters like Mean Residence Time (MRT), flow rate, Residence Time Distribution (RTD), etc. of various processes [6].

To carry out the RTD study, radiotracers are ideal tracers used at the industrial scale without interrupting the normal process and often have no competing alternatives [7,8,9]. At the same time, we can follow the two different phases inside the single reactor during operation [10]. RTD of a single screw extruder was obtained at different screw rpm [11]. The tubes of the digester are also identical with the screw extruders which are used to transport the material toward the end inside each tube.

The performance of the pulp digester is affected by different parameters. For optimum pulping, a fixed bath ratio has to be maintained during pulping which depends on the flow rate of the liquid phase and steam [4]. High axial dispersion is required in the initial phase for better pulping. The complex geometry of the digester and variable viscosity of the liquid phase in each tube makes it very difficult to predict the hydrodynamic behavior. RTD of the same digester has been performed using radiotracer ^{82}Br [5]. In the present study, the radiotracer ^{198}Au as chlorouaric acid was used to trace the liquid phase of the three-tube pulp digester operating under transition flow conditions. The MRT and RTD of the system were determined using suitable flow models.

EXPERIMENTAL SECTION

Materials

Wheat straw and white liquor are two main ingredients to produce the pulp through the soda pulping process. White liquor is an aqueous solution of different chemicals including sodium hydroxide as the main constituent present in the amount of 8.28% (w/w) in water. Other chemicals included sodium carbonate, sodium sulphate, and sodium chloride. These chemicals are present in trace amounts. Wheat straw is an agriculture residue and obtained from farmers in a size of 3-5 cm.

Another key ingredient to carry out the RTD study is radiotracer. The radiotracer ^{198}Au is obtained from the Board of Radiation and Isotope Technology (BRIT), India. The radiotracer ^{198}Au is used in the form of chloroauric acid. Properties of the radiotracers are given in Table 1. A little amount of radiotracer (3-5 ml) diluted in 100 mL of water and used to trace the liquid phase of water.

Experimental procedure

The industrial-scale experiments were performed at SATIA Industries Ltd., Punjab, India. The continuous horizontal digester comprises three tubes, which are connected through a vertical neck. The schematic of the digester is shown in Fig. 1. The helical screw in each tube is used to transport the pulping mixture from one tube to the next. A pump is installed to deliver the white liquor at the inlet of the digester. Radiotracer ^{198}Au was spontaneously injected at the inlet of the liquor feeding pump. The detailed pulping process was described in previous studies [4,5].

A radiotracer follows the liquid phase. The sodium iodide scintillation detectors were mounted outside the tubes of digester to measure the concentration of the tracer at the inlet of the first tube and outlet of each tube. These detectors were connected using wires to the data acquisition system to record the data [5]. The experimental conditions for radiotracer experiments are given in Table 2.

Table 1: Properties of the radioisotope.

Isotope	Half-life	Radiation and energy (MeV)	Chemical form	Tracing of the phase
Gold-198 (^{198}Au)	2.7 day	Gamma: 0.41(99%)	Chloroauric acid	Aqueous and solid

Table 2: Experimental conditions and output parameters for different experiments.

Exp. Tag	Wheat straw feeding rate (m^3/min)	White liquor flow rate (m^3/min)	k	RA (g/l)
E_1	47	0.350	14.0	5.12
E_2	47	0.360	12.9	4.80

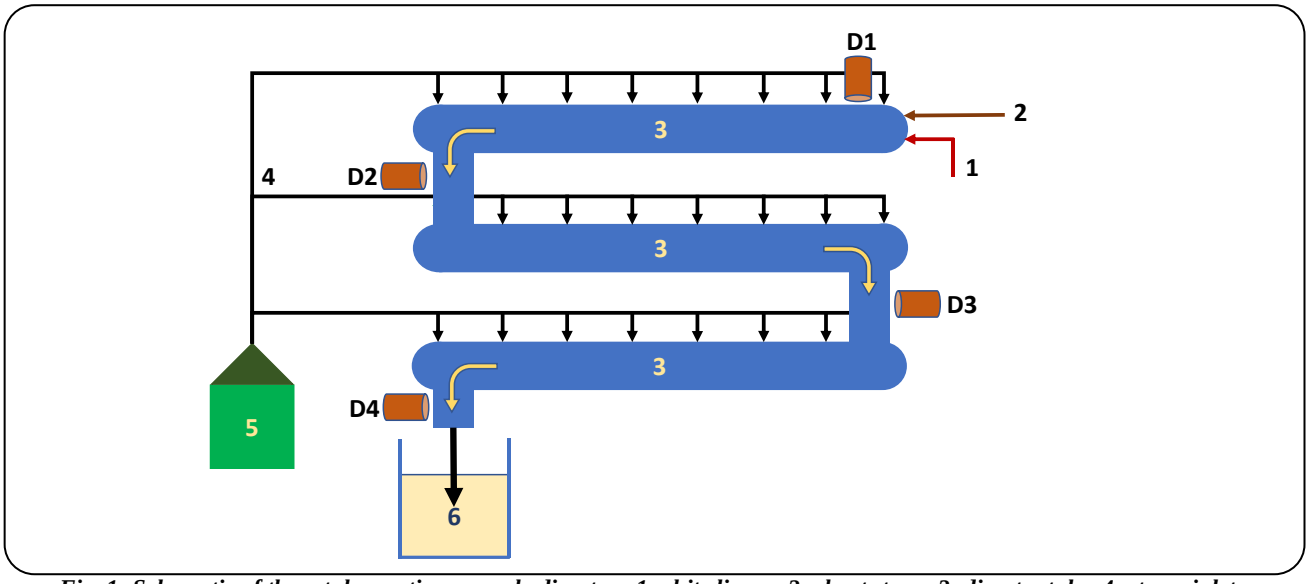

Fig. 1: Schematic of three tube continuous pulp digester: 1-white liquor, 2-wheat straw, 3- digester tube, 4- steam inlets, 5-boiler, 6-blow tank, D1, D2, D3, D4- γ-radiation detectors

Modeling

As the radiotracer passed through the tubes, the gamma energy emitted by the radiotracer is detected by the sodium iodide scintillation detectors mounted on the tubes. The detector measured the strength of gamma energy signals emitted by the radiotracer as a function of time.

The concentration data recorded in the form of signal strength with time and can be represented in the form of C(t) curve. The concentration of the radiotracer is assumed to be uniform in the radial direction and can be represented using the following equation:

$$\frac{\partial C}{\partial t} = -u\frac{\partial C}{\partial z} + D_{ax}\frac{\partial^2 C}{\partial z^2} \qquad (1)$$

The normalized tracer concentration data can be obtained by dividing the concentration by the area under the concentration versus time curve.

$$\text{ormalized tracer concentration} = \frac{C(t)}{\int_0^t C(t)\,dt} \qquad (2)$$

Where the denominator of Eq. 2 denoted the area under the tracer concentration-time curve. Axial dispersion model can be used for turbulent or laminar flow in long tubes, pipes, etc. [6]. There are no limitations for turbulent flow but for laminar flow, the axial dispersion model is applicable only with conditions. Applicability of the axial dispersion model can be detected using Bodenstein number in correlation with the ratio of length to the diameter of the system explained in the literature [6].

In the present case, flow is in a transition phase (nor laminar neither turbulent) for the whole digester. The axial dispersion model with open-open boundary conditions has been chosen for modeling the experimental data. The axial

dispersion model with open-open boundary conditions represented by the following equation is used[6]:

$$E_{ad} = \frac{u}{\sqrt{4\pi(Dt)}} \exp\left[-\frac{(L-ut)^2}{4Dt}\right] \quad (3)$$

Where E_{ad} is the residence time distribution function for the model, u is the velocity of the phase which is being traced, D is the dispersion coefficient, L is the length between two detectors and t is the time variable.

The dispersion of the liquid phase is measured by calculating the dimensionless Peclet number (Pe) which is expressed as below:

$$Pe = \frac{uL}{D} \quad (4)$$

The Peclet number provides information about the dispersion of the liquid phase inside the digester. A low value of the Peclet number represents high dispersion and the high value of the Peclet number represents the plug-flow behavior. At high dispersion, the curve obtains with a sharp peak and slowly decreasing long tail. However, the plug flow behavior is represented by the symmetric or bell-shaped curve [6]. The axial dispersion model is also explained with the convolution process by *Sheoran et al.* [12].

Another model, tank in series with back-mixing is also used to model the experimental data obtained after RTD experiments [13]. The first moment for n number of tanks can be determined by:

$$\tau_n = \frac{1}{N}[n + \beta - (1+\beta)\gamma^k] \quad (5)$$

N is the nth number of tanks connected in series, n is the intermediate tank, β is the backflow ratio, γ is the total flow ratio, k is the cell number from the outlet which is equal to (N-n+1). A similar model with plug flow is used to predict the internal recirculation in a reactor [14].

As the impulse input is injected into the 1st digester tube, the inlet signal for the second and third tube was not ideal due to distortion caused when it passes through first tube. In that case, the convolution procedure is used to obtain the RTD for these tubes. The convolution integral for a discrete signal is given by the following function [6,7]:

$$C_{out}(t_i) = \Delta t \sum_{j=1}^{i-1} C_{in}(t_{i-j}) E(t_j) \quad (6)$$

The concentration was measured every 10 seconds.

The plug-flow component in each case represented the time delay fragment of the concentration data that is, the time before the concentration starts rising. This type of behavior is seen in each case. The plug flow fragment is measured using experimental data simply by measuring the counts before concentration starts rising.

The RTD models (axial dispersion and tank in series with back-mixing model) were directly used for the modeling of the whole digester and first tube. Since the tracer input signal at the feed point was instantaneous. Hence, the tracer signal at the outlet of the first tube could be simulated to an impulse response for the axial dispersion model and tank in series with the back-mixing model. For simulation, the value of parameters is adjusted until the model output fitted the experimental data. However, the above procedure cannot be used for the subsequent tubes. Since there is a non-ideal input signal for the second and third tubes. Hence, the simulation for the second and third tubes has been completed using a convolution procedure. The modeling of experimental data has been done using the software DTSPro V 4.2.

RESULTS AND DISCUSSION

The industrial level RTD experiments were performed for the conditions given in Table 2. A three-tube pulping digester was analyzed for its flow behavior using radiotracer ^{198}Au. The obtained data in concentration versus time was pretreated and normalized for better clarity. The normalized tracer concentration for experiments E_1 and E_2 are shown in Figs. 2 and 3 respectively.

The pulping digester was analyzed in two different ways. One way to consider the whole digester as a single reactor and another way is to assume each tube as an individual/separate reactor. The RTD modeling of all two cases has been performed using the axial dispersion model and tank in series model with back-mixing, each headed by a plug flow component. The convolution approach is used to obtain the non-ideal inputs into ideal inputs for the second and third tubes [5, 7].

Soda pulping of wheat straw is carried out at a 150-170 °C temperature and 5-7 bar pressure in the presence of pulping

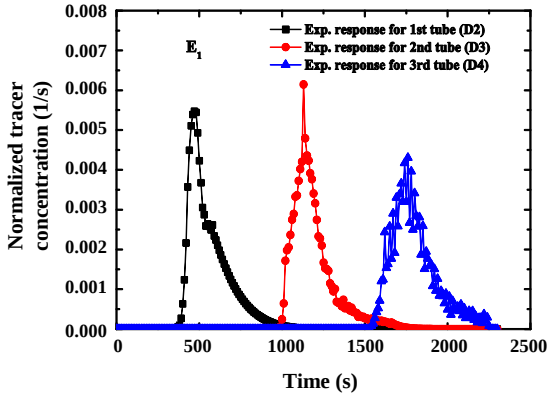

Fig. 2: Experimental data for experiment E_1.

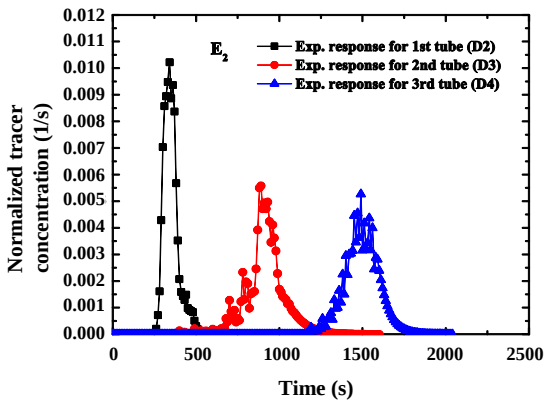

Fig. 3: Experimental data for E_2.

chemicals (white liquor). The wheat straw feed rate was set as 47 m^3/min and the white liquor flow rate was varied from 0.35 to 0.36 m^3/min. The high-pressure steam was continuously fed to the pulping digester to maintain the desired temperature and pressure for high conversion and increased yield.

Inside the first tube of the digester, the liquid phase in the form of white liquor has a viscosity and specific gravity nearly equal to water. Under these conditions, the pulping of wheat straw is started. The pulping of wheat straw occurs at 5-7 kg_f/cm^2 pressure and 155-170 °C. As the material moves from the first tube to the third tube the extent of pulping increases and the heterogeneous mixture of the first digester tube is converted into the homogeneous mixture in the third digester tube. It shows that the dispersion of the liquid phase decreases along the length of the digester tube. The dispersion of white liquor inside the tube of the digester has been determined using the Peclet number (Pe). Pe gives the dispersion of the material or solute alongside the flow's longitudinal direction. The low value of Pe for the first tube shows very high dispersion in the first tube. The model parameters for both experiments are given in Table 3.

The observation of Table 3 shows that the dispersion is increasing as the material passes from the inlet to the outlet of the digester. The maximum value of Pe is observed inside the third tube which represents the plug flow behavior or negligible dispersion inside the third tube. The complete transition of the flow from high dispersion to low dispersion due to an increase in the viscosity of the liquid phase. The viscosity of the liquid phase is increasing along the length of the digester due to the delignification reaction. As the delignification reaction progresses, the dissolved lignin content increases in the liquid phase and leads to the higher viscosity of the liquid phase at digester outlet than the inlet.

The curves obtained at the outlet of the first digester tube (Figs. 2 and 3) show that sudden increases in the concentration and having a long tail for both experiments. This behavior of these curves is representing the mixed flow with high dispersion coefficient at the outlet of the first digester tube. The tank in series with back-mixing model and axial dispersion model is best fitted with the experimental data (E_1 and E_2) and shown in Fig. 4. The model parameters for both models are given in Table 3. It is observed that the dispersion decreases along the length of the digester tube from inlet to outlet.

Furthermore, the whole digester is assumed as a single reactor and the RTD signals at the third tube outlet are considered for the study (Fig. 5). Similar to the first digester tube, the tank in series with back-mixing model and axial dispersion model are best fitted with the experimental data (E_1 and E_2) and the model parameters are given in Table 3. The observation of Fig. 5 shows that the dispersion is further decreased to a minimum value in the third digester tube. The convolution procedure is adopted to convert the no-ideal input of the second and third digester tube into the ideal signals to obtain the RTD of the second and third digester tube, as discussed above in the modeling section [5].

The RTD modeling of the second digester tube is shown in Fig. 6 for both experiments E_1 and E_2 based upon the convoluted input. Again, the tank in series with the back-mixing model and axial dispersion model were found suitable to represent the experimental data.

Table 3: Model parameters for both experiments.

Exp. Tag	Description	Plug flow component (s)	Tank in series with back-mixing model			Axial dispersion model	
			No. of tanks (N)	Back-mixing ratio (α)	τ (s)	Pe	τ (s)
E_1	1st tube	380	4	3.2	180	2	140
	2nd tube	480	17	1.8	140	9	130
	3rd tube	600	61	0.6	120	29	92
	Whole digester	1430	31	2.3	380	16	350
E_2	1st tube	230	7	5.2	68	3.5	45
	2nd tube	470	20	2.3	168	11	180
	3rd tube	500	56	0.9	98	26	110
	Whole digester	1200	26	2.8	360	13	340

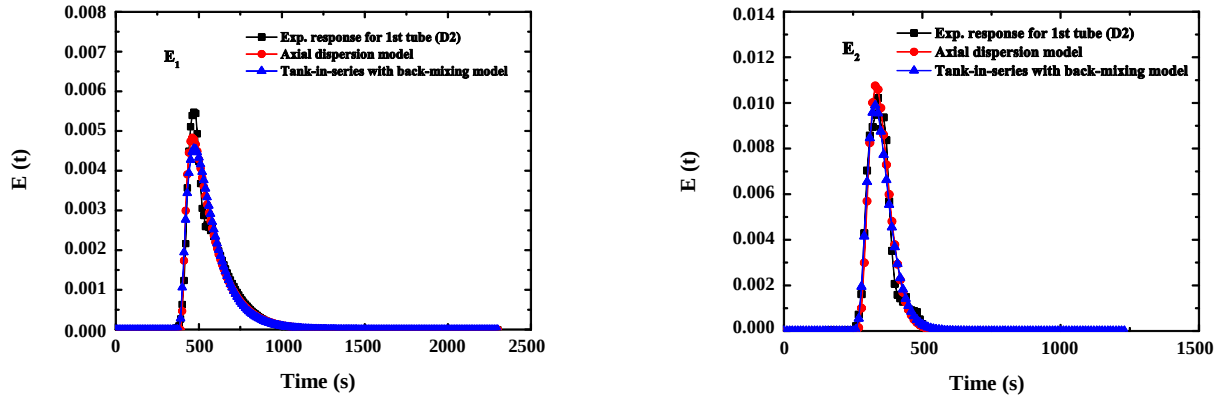

Fig. 4: RTD modeling for first tube for experiments E_1 and E_2.

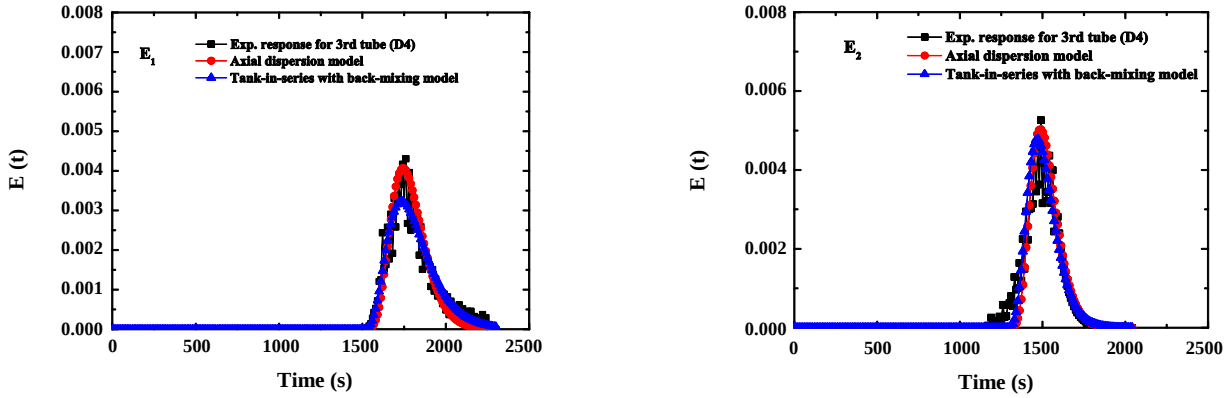

Fig. 5: RTD modeling for the whole digester by assuming it a single reactor for E_1 and E_2.

The model parameters are given in Table 3. The $E(t)$ curves of Fig. 6 are showing the lower dispersion in the second tube than the first digester tube. This is due to the relatively higher viscous pulping mixture than in the first digester tube. At this stage, the delignification process is partially completed and the lignin content is increased in the second tube, which leads to higher viscosity in the second digester tube than the first one.

Overall dispersion inside the digester is almost equal to that represented by the second tube. Similarly, the RTD

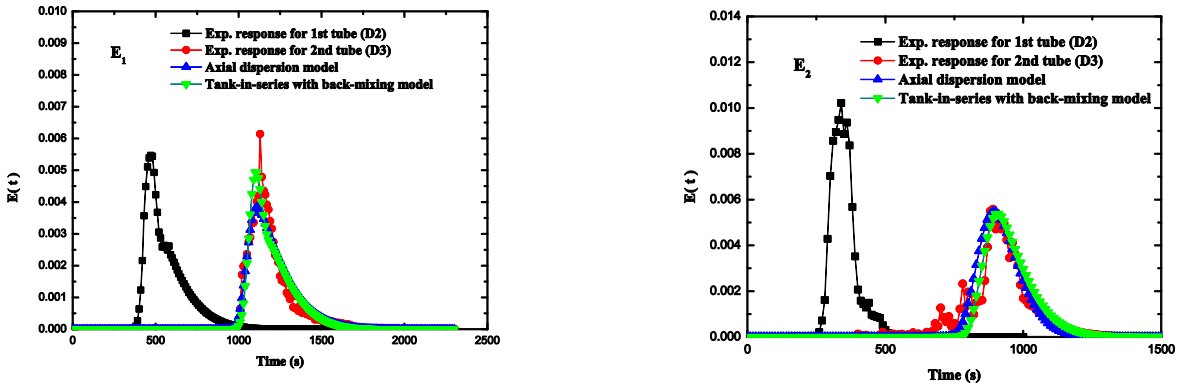

Fig. 6: RTD modeling for second tube using convolution for E_1 and E_2.

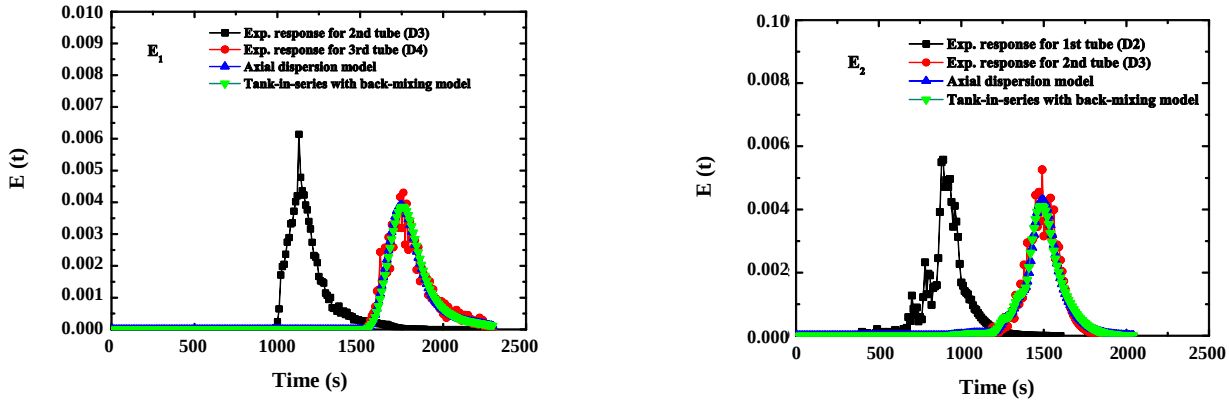

Fig. 7: RTD modeling for the third tube using convolution for E_1 and E_2.

modeling of the third digester tube is shown in Fig. 7 for both experiments E_1 and E_2. The model parameters for the tank in series with the back-mixing model and axial dispersion model are given in Table 3. Fig. 7 shows the symmetric curve behavior, which is showing almost negligible dispersion in the third tube than the first digester tube. This negligible dispersion refers to the plug flow behavior inside the third digester tube. This is again due to the highly viscous material in the third digester tube than the first and second tube. In this tube, the delignification process is completed and the lignin content is maximum, which leads to the highest viscosity in the third digester tube.

The predicted Mean Residence Times (MRTs) of the three tube pulp digester is shown in Table 4. The tank in series with a back-mixing model and axial dispersion model was used to obtain the MRTs of the whole digester and individual tubes (Table 4). The observation of Table 4 shows that the MRT of the first tube is the lowest for both experiments E_1 and E_2. Inside the first tube, the low viscosity of the pulping mixture results in high dispersion, and a high space velocity of liquid results in high back-mixing. As the viscosity increases inside the second tube, the space velocity reduces, and low back-mixing is observed. In the third tube, a further increase in the viscosity leads to the transformation of flow into plug flow behavior. Very low back-mixing was observed in the third tube.

The liquid inlet flow (Table 2) is higher in experiment E_2 than E_1, which leads to a higher space velocity of liquid in the case of E_2. Hence, the total MRT is lower in experiment E_2 than E_1 due to the high white liquor flow rate.

Table 4: Mean residence time for each tube and whole digester.

Exp. Tag	Description	Mean residence time (MRT) (min)	
		Tank in series with back-mixing model	Axial dispersion model
E_1	1st tube	9.3	8.6
	2nd tube	10.3	10.2
	3rd tube	12	11.5
	Whole digester	30.1	29.6
E_2	1st tube	5.8	5.6
	2nd tube	10.6	10.8
	3rd tube	9.9	10.1
	Whole digester	26	25.6

Furthermore, the observation of Table 2 shows that the number of tanks (N) in the tank in series with the back-mixing model is almost double the value of the Peclet number (Pe). It may show that the axial dispersion model is representing the plug flow stream overlapped with some amount of back-mixing. The two separate phases converted into the pulp slurry at high temperature and pressure which flows like a plug flow in the third tube. From the hydrodynamic point of view, a transition flow behavior is observed for the whole digester. The concept of transition flow can be applied only for the case when we assumed the whole digester as a single reactor. For this case, we assumed that viscosity is constant at each point in the digester which is equal to the viscosity of the pulp. But in actuality, the viscosity and specific gravity are continuously changing from the inlet to the outlet during the delignification process. As the first tube of the digester having a high temperature and pressure, the cellulose fibers are started to separate from lignin. The lignin gets dissolved in the liquid phase which resulted in a high viscosity of the liquid phase. Also, the dissolved lignin and other chemicals turned the liquor black. Inside the third tube, the liquid phase has a high viscosity and density with plug flow behavior.

CONCLUSIONS

The radiotracer ^{198}Au was successfully used to follow the liquid phase in the industrial scale continuous pulp digester and found suitable to the RTD measurement of the industrial scale pulp digester. The radiotracer ^{198}Au was injected at the digester inlet to follow the liquid phase of the digester. The axial dispersion model and tank in series with a back-mixing model preceded by a plug flow component were found appropriate to simulate the experimental RTD data. These models are also found suitable to predict the inside flow behavior of the pulp digester. High dispersion was observed in the first tube, which is decreasing in the subsequent digester tubes and finally converted into plug flow in the third digester tube. Overall, the transition flow behavior is observed for the whole digester with a large back-mixing in the first tube, again this back-mixing is decreasing along the length as we move toward the third tube outlet. The MRTs of the whole digester and individual tubes were also measured.

Acknowledgments

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Nomenclature

ADM	Axial dispersion model
C	Concentration of the tracer, counts/s
D	Dispersion coefficient, m^2/s
Di	Detectors (i = 1, 2, 3..)
DAS	Data acquisition system
E_{ad}	Exit age distribution of the axial dispersion model, s^{-1}
k	Kappa number
L	Length between two detectors, m
MRT	Mean residence time
N	Number of tanks
Pe	Peclet number
RA	Residual alkali, g/L
RTD	Residence time distribution

TISBM Tank-in-series with back-mixing ratio
t Time, s
u Velocity of the liquid phase inside the digester, m/s
z Length of the reactor

Greek symbols

τ Mean residence time, s
β Back-mixing ratio in tank-in-series with back-mixing model
γ Total flow ratio in tank-in-series with back-mixing model

Subscripts

ad Axial dispersion model for large extent of dispersion
i 1,2,3…….
in inlet
out outlet

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