Ms Meenakshi Sheoran

Dr. Avinash Chandra

Dr. Haripada Bhunia

Dr. Pramod K. Bajpai

Dr. Harish J. Pant
and
Mr. S. Madhukar Rao

SATIA INDUSTRIES LTD.

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INVESTIGATION THE FLOW BEHAVIOR OF CONTINUOUS PULPING DIGESTER USING RADIOTRACER TECHNIQUE TO OPTIMIZE THE OPERATING CONDITIONS

Abstract

The modern pulp and paper mills are shifting from wood based raw material to alternate cellulose rich raw material such as agro-residues like, wheat straw, bagasse, etc. It is due to rapidly shrinking forest fields and other environmental concern. This shift made the pulping process as continuous process rather than the traditional batch operation, hence, it is facing new technological challenges. In the continuous process, the raw material is being digested in horizontal continuous pulping digester to obtain pulp. The output quality of the produced pulp is highly influenced by the time spent by the raw material inside the digester in the presence of white liquor at high temperature and pressure. In this regard, an industrial scale three tube continuous pulp digester has been analyzed using latest non-destructive radiotracer residence time distribution (RTD) technique. The radioisotope ^{82}Br (γ energy source) has been used as radiotracer to trace the liquid phase of the pulp digester. The biomass flow rate and white liquor flow rate are considered as the process variables for the experiments. The obtained RTD data was treated and used to explain malfunctioning and flow abnormalities in the pulp digester. Flow channeling was observed in the first tube of the digester. The mean residence time (MRT) of pulp digester is also estimated for various sets of conditions. The influence of MRT on the pulp quality has been correlated with Kappa number (***k***) and residual alkali. Based on the obtained results and analysis, the optimum operating conditions have been recommended for the digester.

Keywords*:** Pulp digester, Kappa number (k***), Mean residence time, Radiotracer, axial dispersion.

1. Introduction

The Indian paper industry is among the top 12th industrial sector in the world with an approximate paper production of 13.5 tons annually. The estimated turnover of the Indian paper industry is approximately Rs. 4000 crore. In India, the demand of paper is increasing day by day at a rate of ~5.5 % per annum and the estimated demand in 2025 would be over 22 million tons. The paper mills in India are aiming to achieve the projected demand of 22 million tons by the year 2025 and many of them are in the spree of upgrading the plants with modern technologies. The paper industry in India is on growth trajectory and is expected to touch 7.5 % GDP in coming years [1]. Different paper products produced in paper mills are based on different types of raw materials. These raw materials contain cellulose, hemicellulose and lignin in different composition depending on the type of biomaterials. There are mainly two processes for pulping, namely mechanical and chemical. In mechanical process, lignin is separated by mechanical disintegration, produced high strength paper but pulp contain high lignin content which is the reason of yellowish paper products [2]. In chemical pulping process, chemicals are used to separate the lignin from cellulose. Lignin is a complex material which binds the cellulose fiber and acts like glue. During delignification reaction, lignin cannot be removed completely because the chemical used for pulping also degradethe cellulose and hemicellulose fibers and make them weaker which further influencethe pulp yield and fiber quality. To achieve the acceptable pulp yield and fiber quality, delignification reaction has to control and stopped when the lignin content was low enough for fiber separation [2].

For better control over delignification process, the process variables have to be controlled. Due to different types of feed stoke used for pulping, their process variables are also difficult to measure. The important quality measurement method is the ***k*** number which indicates the degree of delignification. This ***k*** number is directly related to the extent of reaction. The phenomena in this process are more complex and are a mixture of bulk flow, diffusion, convection etc. [2].

Every pulping industry wants to optimize the pulping process by minimizing the variation in the ***k*** number.The optimized conditions can increase the product yield, quality, reduced waste and operating cost etc. These characteristics can be obtained by removing the malfunctioning in the digester and improper operating conditions. These malfunctioning and flow behavior of any reactor can be described by fluid mechanics and residence time distribution (RTD) fundamentals. RTD is the probability distribution function of the material flowing inside the reactor [3]. By measuring RTD, we can estimate the efficiency of the process, mixing time, bypassing, channeling, dead zone etc. [4]. The improper design of the reactor can be optimized at the designing stage by determining the hydrodynamic behavior at the different operating conditions. A number of studies have been done to measure the flow rate of gases, solids and liquids in different industries using RTD [4-7]. The tracers are used to carry out the RTD test by tracing the process material inside the reactor. A number of conventional tracers like dye, salts, fluorescent materials, electrolytes, ions etc. are used as tracer but radioisotopes have number of advantages over conventional tracers [5]. Radiotracers are useful in opaque system, have high detection sensitivity, continuous sensing, wide range of availability and physical/ chemical compatibility with the process materials [6]. Due to their radioactive decay, the radiotracer disappears from the tagged system with time and the contamination due to radioactivity removed automatically. Radiotracer based technology is rapidly accepting in various applications / process in industries to optimize the process variables and understand the flow kinematics. The radiotracer based RTD technology covers wide range of applications in various industries like petroleum, mineral processing, petrochemicals, pulp and paper, waste water treatment etc. [4, 7-13]. The radiotracer application includes leak detection, material bypassing, channeling, recirculation, blockage detection, flow rate measurement, mixing time measurement, etc. [14]. In our recent study [7], the radiotracer based RTD technique was used to investigate the continuous pulp digester using ^{82}Br. Overall, there is only our previous study is available in literature on RTD measurement of pulp digester.

The presented work is the extension of our published work [7]. The radiotracer experiments were performed at SATIA Industries Ltd., Muktsar, Punjab. The present work is showing the feasibility of the radiotracer RTD experiments at industrial scale without disturbing the running processes. Hence, in this work, radiotracer experiments were carried out in continuous horizontal three tube pulp digester using the trace amount of ^{82}Br as a radiotracer material for tracking the liquid phase.

2. Material and methods

Wheat straw is a complex mixture of lignin, cellulose and hemicellulose as three main components, and a small amount of soluble substrates (also known as extractives) and ash like as other biomass of ligno-cellulosic composition. The overall chemical composition of wheat straw might slightly differ depending on type of wheat species, soil, climate and cultivation conditions. The wheat straw contains ~32-40% w/w cellulose, ~20-25% w/w hemicellulose and ~15–20% w/w lignin.

The other raw material used for pulping is white liquor. White liquor is the solution of various chemicals used for pulping process. The sodium hydroxide is the key component of the white liquor and is present in ~8.28% (w/w) in the solution. To prevent the degradation of the cellulose fiber, sulfur as sodium sulfite is used in very small quantity ~0.44% (w/w). Sodium carbonate is also present in trace amount ~0.88% (w/w) in the white liquor whereas the sodium chloride is present in ~2.20% (w/w). All the chemicals used in the preparation of white liquor were manufactured by SATIA Industries Ltd., Mukatasar, Punjab, itself.

One of the bromine isotope, ^{82}Br is the radioactive substance having half-life of 36 hours with gamma energy 0.55 MeV (70%). The ^{82}Br in the form of ammonium bromide was used as a radiotracer material for these investigations. ^{82}Br was obtained from Board of Radiation & Isotope Technology (BRIT), Mumbai, under the supervision of a team of scientists from Bhabha Atomic Research Center (BARC), Mumbai.

2.1 Experimental procedure

A pulse experiment technique was used for the determination of the residence time distribution for the three tube continuous pulp digester. A series of radiotracer experiments were carried out at different flow operating conditions shown in Table 1. The small amount (2-5 ml) of the ^{82}Br (depending upon the activity of the radionuclide at the time of the experiment) was dissolved in 100 ml of water. This solution was instantaneously injected in the white liquor inlet pipe near to the feed point of the digester, and then it moved along with the pulping mixture in the digester. Four detectors D_1, D_2, D_3 and D_4 have been placed to trace the radioactive material ^{82}Br as shown in figure 1. A multi-channel data acquisition system (DAS) has been used to record the online data. These detectors were connected to the DAS using wired network. The DAS was set to record data points with a sampling time of 10 sec. As the radioactive material passes from the detectors D_1 to D_4, a pulse has been observed on the monitor and recorded in the computer connected to DAS. Initially at detector D_1, it shows very sharp peak with high energy. Further, as the material moves along the digester from detector D_1 to D_4 the peak height reduces (energy reduces) and broadness of the peak increases.

2.2 Data analysis

The detector recorded the γ-energy as a function of time at the four positions of the digester. The recorded data were post-processed to obtain the meaningful data-set, which may able to understand the underlying physics. The post processing includes, background correction, radioactive decay correction and zero shifting.

The injection of the tracer should be fast to obtain the pulse input. Assuming $C(t)$ is the energy concentration of the radiotracer at time, t. At $t = 0$, the radiotracer was injected as pulse input function and a sharp peak was observed at detector D_1 (Figure. 2). The subsequent peaks were observed in detectors D_2, D_3 and D_4 with increase in time ($t > 0$) (Figure. 2). The obtained RTD function $E(t)$ can be defined as [3,7]

$$E_i(t) = \frac{C_i(t)}{\int_0^\infty C_i(t)dt} \quad (1)$$

Such that $$\int_0^\infty E_i(t)dt = 1 \quad (2)$$

Where i = 1, 2....n, and n is the number of points.

The simple RTD data treatment is the calculation of moments. The moments are used to characterize the RTD function in terms of statistical parameters such as mean residence time and standard deviation. The first moment around the origin is determined using the following relation: [11]

$$M_i = \int_0^\infty t\ E(t)dt \quad (3)$$

Where, i = 1 for input and i = 4 for output.

The mean residence time ($\bar{t}$) for the whole digester is defined as follows:

$$\bar{t} = M_4 - M_1 \quad (4)$$

And the mean residence time for individual tube is given as:

$$\bar{t}_j = M_{j+1} - M_j \quad (5)$$

Where j is representing the digester tube 1, 2 and 3. The detailed data analysis procedure is given in our previous work [7].

The theoretical mean residence time for digester having volume V with average volumetric flow rate f is defined as [3].

$$\tau = \frac{V}{f} \qquad (6)$$

3. Results and discussion

The radiotracer RTD experiments were performed at various operating conditions given in Table 1. The two different feed screw speeds at 55 rpm and 65 rpm were chosen for these sets of experiments.The obtained RTD data are plotted and shown in Figure 2 for different sets of operating conditions (Table 1).

In the first set, run 1 and run 2 (Table 1) were carried out at a screw speed of 55 rpm and in second set of experiments, run 3 and run 4 (Table 1) were carried out at screw speed of 65 rpm. In each case, the radiotracer was monitored at inlet and outlet of the each digester tube (D_1 – D_4). A very sharp peak with negligible width was observed at inlet (D_1) of the first tube of the digester and the peak gets broadened as material moves from inlet to outlet of first tube (D_2), then second (D_3) and third tube (D_4). For each RTD curves shown in Figures 2, the experimental mean residence time were determined and tabulated in Table 2.

The delignification process involves three steps, which are absorption, bulk delignification and residual phase delignification. During the digestion process, the white liquor is absorbed by the wheat straw which is the reason behind consumption of huge white liquor in first tube. At high temperature and pressure the lignin starts to dissolve in white liquor. A little amount of delignification occurs in tube 1 and bulk delignification occurs in digester tube 2. The final cooking phaseand/or finishing steptake place in tube 3. Additionally, the partial degradation of carbohydrates and cellulose fibersare also occurring in tube 3. The degradation of carbohydrates and cellulose fibersin tube 3 is adverse because it diminishes the pulp yield and fiber properties [2], which cause to lower grade paper production.

Four experiments were presented herefor the various operating conditions shown in Table 1. The pulp digester output parameters k values and residual alkali were also tabulated in Table 1. The observation of Table 1 shows that the complex dependencies of k values and residual alkali on operating conditions. It was observed that as the feed screw rpm increases from 55 to 65, the k value decreases whereas the white liquor feed rate increases, the k value decreases only for 55 rpm feed screw speed. The increase in the white liquor feed rate for 65 rpm feed screw speed remain ineffective. It might be showing the saturated amount of white liquor at 65 rpm.The theoretical mean residence time was calculated for each run and shown in Table 2.

In run 1, For the 55 rpm screw speed, and 300 l/min, the tracer material crossed the detector D_1 in few seconds. The tracer starts crossing detector D_2 after about 6 min, a broader peak is observed by detector D_2. The broadness of peak further increases while radiotracer crossing the detector D_3 and D_4. The mean residence time (MRT) for this case was estimated as 28.3 min. which is smaller than the theoretical MRT. Small fluctuations were observed in the RTD curve at tube 2. It also shows the radial recirculation of the pulp mixture. Further, observation of the Figure 2 shows that the axial dispersion of pulp material is decreasing from digester tube 1 to tube 2 and tube 2 to tube 3. At the outlet of the digester tube 3, the pulp mixture is behaves like plug flow.

In run 2, the white liquor flow rate was increased by 10 l/min. (i.e., 310 l/min) by keeping screw speed 55 rpm. The RTD curve for this case is shown in Figure 2. In this case, the radiotracer took 6.1 min to cross the first tube of the digester which is 2 min earlier than the previous case (run 1). The higher flow rate of white liquor increases the average velocity of pulp mixture which enhances the axial dispersion. The radiotracer took ~9.9 min and ~10 min to cross the second and third tubes of the digester respectively. The total time taken by radiotracer to reach at exit point of the digester is 26 min. and for the same case the value of theoretical MRT is 33 min which is much higher than the experimental MRT. This indicates the bypassing and channeling of liquid in pulp digester but no flow abnormalities were seen in figure 2.

For next set of experiments (run 3-4), new conditions were set i.e., the screw speed was set as 65 rpm and white liquor flow rate varied from 355 l/min to 365 l/min The theoretical MRT for screw speed 65 rpm is 29.4 min.

For run 3, the RTD plot is shown in Figure 2 with overall experimental MRT of 24.7 min. which is much lower than the theoretical MRT. The tracer crossed the first, second and third digester tubes in 6.3 min, 7.9 min and 10.5 min. respectively. The obtained RTD curves for this case show normal behavior of the digester with little bypassing and channeling.

In the next run (run 4), the white liquor flow rate is further increased to 365 l/min as shown in Table 1. The RTD plot for first digester tube (D_1) is broad and short in height. The RTD curve for D_2 is showing double peaks at the outlet of the first tube of the digester which was the indication of channeling in the first tube of digester. The tracer passed through the second and third digester tubes in 4.3 and 5.2 min. respectively. The total MRT of the experiment was 34.5 min. which is higher than the theoretical value.

The RTD curve (Figure 2) for run 1, run 2 and run 3 are showing the common flow behavior as axial dispersion. Whereas, the run 4 shows double broaden peak for D_2 at the outlet of first digester tube. This response on D_2 shows the lumping and stucking of the biomass inside the digester tube 1. Two peaks shows the two biomass lumps are passing through the detector D_2 at different time. Further, the broadening of peak can interpret as the size of the lumps. At high white liquor flow rate, bypassing and mixing may appear and at low white liquor flow rate, digestion was not proper.

The ***k*** number is the measurement of the extent of delignification of the lignocellulosic biomass and it is determined using the acidified potassium permanganate solution [15]. Similarly, residual alkali (RA) is estimated after pulping process, by titrating against hydrochloric acid of known strength [16]. A balance need to be maintained between the ***k*** and RA for effective digestion process. The values of the ***k*** and RA were estimated for different experimental conditions and are tabulated in Table 1.

In first set of run (run 1-2),at screw speed of 55 rpm, the values of ***k*** are observed as 14.2 and 13.4 for the equivalent white liquor flow rates of 300 and 310 l/min respectively. The k decreases with gradual increase in white liquor flow rate from 300 to 310 l/min. Conversely, the observation of Table 1 shows that the amount of RA also increases in product stream with increasing the white liquor flow rate.

In the second set of the experiments (Run 3-4), with 65 rpm screw speed, the k showed similar trend. The kin experiment 3 and 4 is 12.8. Furthermore, in experiments 4, RA decreased to 5.01 g/l.

Based on above discussion, the run 3 is found optimum case with the smallest value of k. The value of residual alkali and MRT for this optimum case are 5.33 g/l and 24.7 min. respectively.

4. Conclusion

The RTD analysis technique was used to analyze the industrial scale continuous three tube digester. The radioisotope ^{82}Br was used as a tracer for liquid phase without disturbing normal pulping process. The operating conditions were assumed as process variables and these conditions are optimized using radiotracer RTD technique. At low white liquor flow rate, digestion was not proper which results in high k and high value of residual alkali. At higher white liquor flow rate, channeling and back-mixing was appear in the digester tubes. The operating conditions of run 3 was identified as optimum operating conditions for three tube continuous pulp digester based on obtained value of ***k*** number and residual alkali.

The radiotracer technique may also suitable for detection of the plugging /chocking in the process vessel. In case of plugging, the radiotracer will stuck with the plug material and will give continuous signal of same intensity from one location and unexpected increase in MRT can be seen. It will also replicate in signaling pattern of RTD. The extent of uniform cooking can interpret from the shape of RTD curve. The high sharp peak will show the uniformity for cooking while the short and broaden peak will represent the distributed cooking. Additionally, the comparison of the instantaneous MRT with the optimum MRT may also provide the cooking information as, if instantaneous MRT < optimum MRT, the pulping material will be under cocked and vice-versa.

Overall, this RTD investigation shows the feasibility of the radiotracer RTD experiments at industrial scale without disturbing the normal operation / process and this technique is suitable for optimizing the process variable.

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TABLE
OPERATING CONDITIONS FOR DIFFERENT RUN

Run No.	Screw speed (RPM)	Liquor flow rate (l/min)	Steam Flow (Ton/hr).	Steam Pressure (kg/cm^2)	Tube 1 Temp (^oC)	Tube 2,3 Temp (^oC)	Kappa number (***k***)	Residual alkali *RA* (g/l)
Run 1	55	300	15.00	6.65	157	162	14.2	4.60
Run 2	55	310	18-19	5.98	167	165	13.4	6.14
Run 3	65	355	16	6.26	165	163	12.8	5.33
Run 4	65	365	17	6.32	160	166	12.8	5.01

TABLE
MEAN RESIDENCE TIME (MRT) FOR PULP DIGESTER

Run No.	Theoretical MRT (min.)	Experimental MRT (min.)
Run 1	33	28.3
Run 2	33	26
Run 3	29.4	24.7
Run 4	29.4	34.5

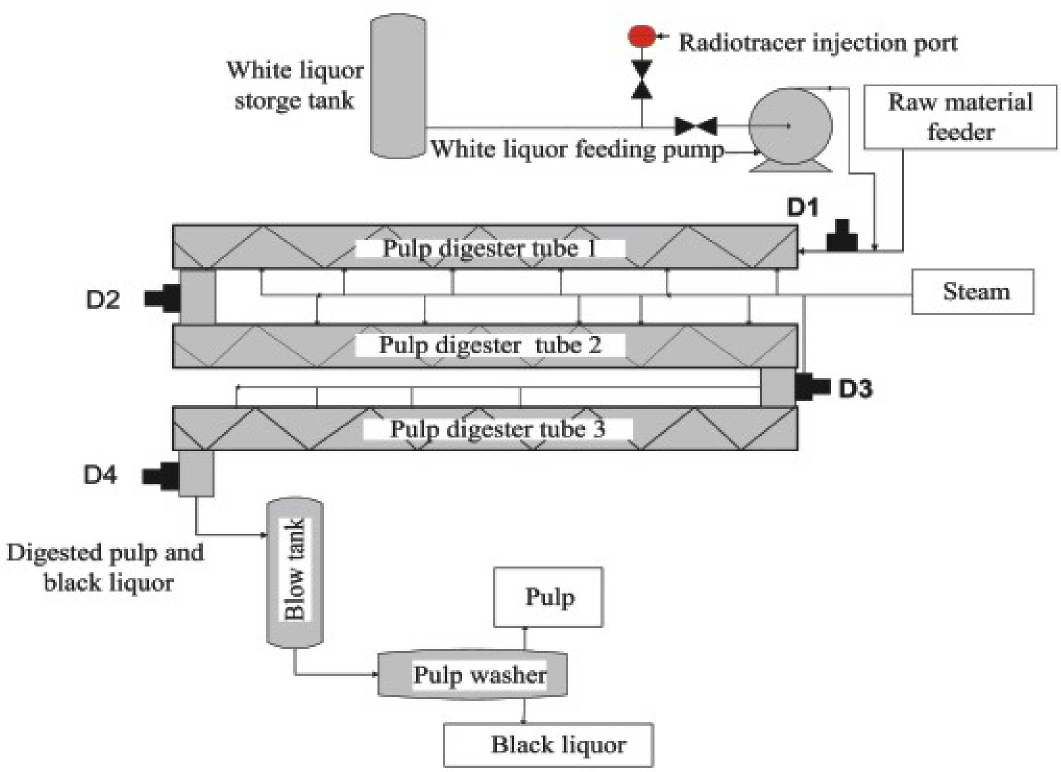

FIGURE SCHEMATIC DIAGRAM AND EXPERIMENTAL SET UP FOR CONTINUOUS DIGESTER

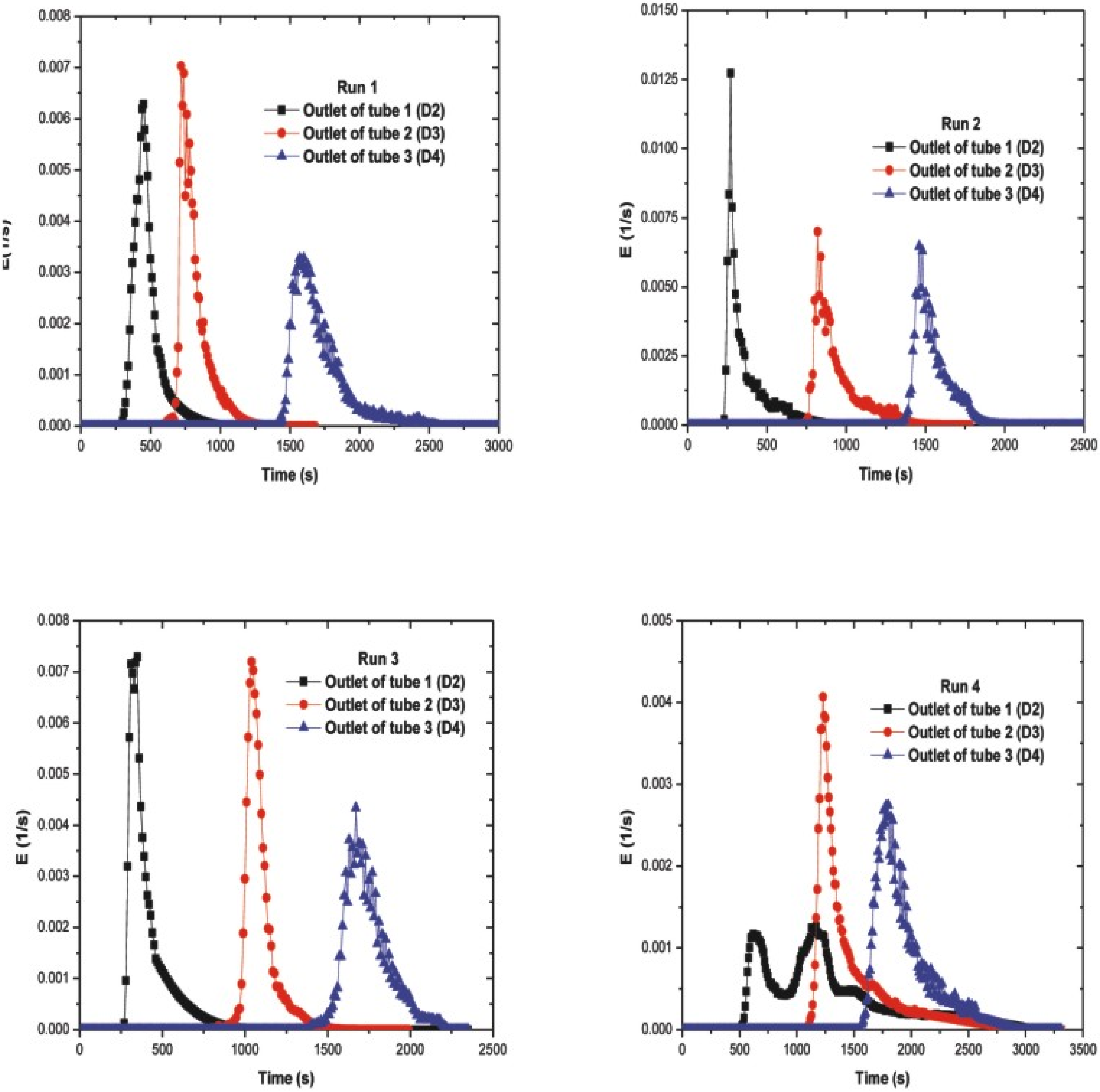

FIGURE NORMALIZED TRACER CONCENTRATION CURVES FOR DIFFERENT RUN

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