



# Radiotracer investigation of a pulp and paper mill effluent treatment plant

Metali Sarkar,  
Vikas K. Sangal,  
Haripada Bhunia,  
Pramod K. Bajpai,  
Harish J. Pant,  
Vijay K. Sharma,  
Anil Kumar,  
A. K. Naithani

**Abstract.** The pulp and paper industry is highly dependent on water for most of its processes, producing a significant amount of wastewater that should be treated to comply with environmental standards before its discharge into surface-water reservoirs. The wastewater generated primarily consists of substantial amounts of organic, inorganic, toxic and pathogenic compounds in addition to nutrients, which are treated in an effluent treatment plant that often combines primary, secondary, tertiary and advanced treatments. However, the treatment methods vary from industry to industry according to the process utilized. The effective performance of effluent treatment plants is crucial from both environmental and economic points of view. Radiotracer techniques can be effectively used to optimize performance and detect anomalies like dead zones, bypassing, channelling, etc. in wastewater treatment plants. Experiments on the distribution of residence time were performed on the aeration tank and secondary clarifier of a full-scale pulp and paper mill to study the flow behaviour as well as locate system anomalies and hence evaluate the performance of the treatment plants using the radiotracer I-131. The convolution method was applied to model the system with an imperfect impulse radiotracer input. The aeration tank was working efficiently in the absence of any dead zones or bypassing. Various hydrodynamic models available in the literature were applied on the aeration tank and secondary clarifier to obtain the hydraulic representation of the systems.

**Keywords:** aeration tank • secondary clarifier • residence time distributions RTD • radiotracer • convolution

M. Sarkar, V. K. Sangal✉, H. Bhunia, P. K. Bajpai  
Department of Chemical Engineering,  
Thapar Institute of Engineering and Technology,  
Bhadson Road, Patiala, Punjab 147004, India,  
Tel.: +91 981 501 5705,  
E-mail: vksangal@gmail.com

H. J. Pant, V. K. Sharma  
Isotope Production and Application Division,  
Bhabha Atomic Research Centre,  
Mumbai, India

A. Kumar, A. K. Naithani  
Shreyans Industries Ltd.,  
Village Malikpur, Sangrur District, Ahmedgarh,  
Punjab 148021, India

Received: 21 April 2017  
Accepted: 7 November 2017

## Introduction

The Indian pulp and paper industry is a major consumer of water, utilizing around 60–120 m<sup>3</sup> of water per ton of paper produced, depending on the type of process used for paper production [1]. The process of manufacturing paper and pulping generate tons of wastewater that contains large quantities of pollutants which are unfit to be directly disposed of in surface-water reservoirs. Wastewater from the pulp and paper industries contains a significant amount of organic, suspended solids, nutrients and occasionally toxic compounds [2, 3]. However, the organic load of this industry requires special attention as it causes the maximum amount of pollution and has widespread implications for what is exposed to it [4, 5].

The activated sludge process (ASP) is one of the most commonly used processes for the biological treatment of dissolved organics in the pulp and paper industries. The aeration tank, used widely for the removal of dissolved organic compounds in the wastewater, is the most crucial part of the ASP. It is essentially a bioreactor that employs active microorganisms suspended throughout the tank. The

microorganisms breakdown the dissolved organic pollutants of the effluent into biomass. As the degradation of organic pollutants is an aerobic process, diffusers and mechanical aerators are employed in the aeration tank to maintain the desired level of dissolved oxygen required for the process. The biomass formed in the aeration tank is separated from the treated water in a secondary clarifier that acts as a settling tank and works according to the principle of gravity [6, 7]. The hydrodynamics of the aeration tank and secondary clarifier vary greatly from each other, however, maintaining proper hydrodynamic conditions in the reactors may help to combat several flow anomalies like bypassing, dead zones, etc., resulting in the desired treatment of the wastewater to ensure the reactors function properly.

The RTD analysis technique has been extensively used to characterize system performance by identifying possible anomalies present in reactors [8–12]. This is achieved by injecting a minute amount of tracer into the inlet of the reactor and monitoring the outlet. Although several chemical, optical and radioactive tracers have been employed to investigate RTD in wastewater treatment plants, properties like high detection sensitivity and high selectivity, which facilitate *in-situ* measurements and the online collection of data, make radiotracers a superior choice in terms of the RTD analysis of large-scale industrial systems. The advantages of using radiotracers in RTD studies of full-scale industrial processes have been discussed in detail by many researchers [10–19].

The input method for radiotracer injection in the system is pulse input which provides a RTD curve that can be conveniently modelled using the RTD models present in the literature [20, 21]. However, industrial systems are interconnected and the output radiotracer signal from one system acts as the input to a subsequent system. These inputs are non-ideal impulse signals that cannot be directly evaluated and can only be studied using numerical convolution along with the preferred RTD model. The effluent treatment plant at Shreyans Industries Ltd. in India employs a two-stage ASP process that includes two sets of aeration tanks and secondary

clarifiers connected in series, as seen in Fig. 1. Radiotracer experiments were conducted to study the performance and efficiency of ASP. In an earlier study our group reported the hydrodynamics of the first aeration tank and clarifier both with perfect impulse tracer inputs [22]. This paper reports the RTD data from the second radiotracer experiment performed on the second set of an aeration tank and secondary clarifier evaluated using the convolution of input signals.

## Process description

### Experimental setup

The experiment was conducted on the second treatment unit of the effluent treatment plant at Shreyans Industries Ltd. in Ahmedgarh, India. The schematic diagram of the radiotracer experiment at the effluent treatment plant is shown in Fig. 1. The dimensions and process parameters of each system are outlined in Table 1. In the present study, the RTD experiment performed on aeration tank 2 and secondary clarifier 2 is discussed. The aeration tank is 45 m long, 16 m wide and 4.5 m deep with a capacity of 3240 m<sup>3</sup>. The aeration tank was equipped with a diffused aeration system fitted with a fine bubble ethylene-propylene-diene monomer (EPDM) membrane to sparge dissolved air. The effluent feed rate in the aeration tank was 5.21 m<sup>3</sup>/min. The secondary clarifier, with a capacity of 890 m<sup>3</sup>, is a circular settling tank with a central feed of 18 m in diameter and a height of 3.5 m.

### Radiotracer distribution experiment

The RTD experiment was conducted on the full-scale ASP to trace the aqueous phase (wastewater) in the aeration tank and secondary clarifier. The radiotracer iodine-131 (half-life: 8 days, gamma energy: 0.36 MeV) was used as sodium iodide (NaI) which was supplied by the Board of Radiation and Isotope Technology (BRIT) in Mumbai, India. A minute amount, approx. 10 ml, of iodine-131 was dissolved

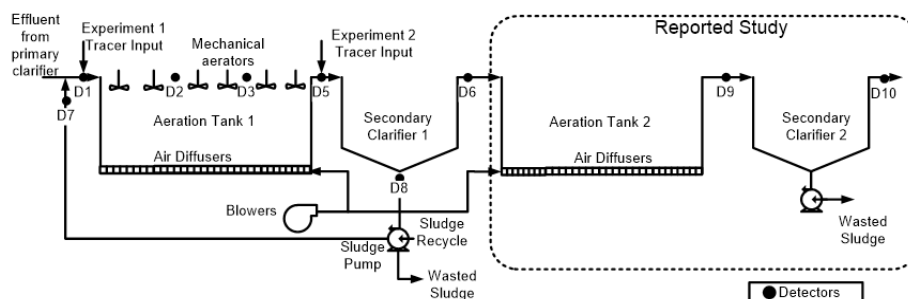


Fig. 1. Schematic diagram (front view) of the complete industrial ASP setup.

Table 1. Process description and calculated parameters

	Dimensions	Volume [m <sup>3</sup> ]	Flow rate [m <sup>3</sup> /min]	Retention time
Aeration tank 2	45 × 16 × 4.5 m	3240	5.21 ± 1%	622 min (10.4 h)
Secondary clarifier 2	d = 18 m, h = 3.5 m	890	5.21 ± 1%	171 min (2.8 h)

in 2 liters of water (activity: 30 mCi in a volume of 5 ml), injected into the inlet of the secondary clarifier, and monitored at the inlet and outlet of the second aeration tank (D9) and secondary clarifier (D10) as shown in Fig. 1. The input signal for aeration tank 2 was obtained by convoluting the imperfect impulse input signal of secondary clarifier 1. The absorption of I-131 by solid sludge was minimal and did not interfere with the RTD study of the aqueous phase in effluent treatment plants [23, 24]. Scintillation (NaI) detectors were placed at the inlet and outlet streams of the system and were connected to a data acquisition system (DAS). The DAS was linked to laptops and the sampling time to record data points was set at 5 min. The duration of the experiment was 50 hours, which was approximately 3 times longer than the theoretical/expected mean residence time, to obtain a proper description of flow.

### Data treatment and analysis

#### Calculation of theoretical and experimental MHRT

The RTD data were recorded in terms of counts per second from the outlet stream of each system and pre-treated to eliminate experimental errors. The RTD curves were normalized to obtain a graph that plots  $E(t)$  against time. The theoretical mean hydraulic retention time (MHRT) of the systems were calculated using Eq. (1) [10, 11, 20, 21].

$$(1) \quad \tau = \frac{V}{Q_0}$$

where,  $V$  – volume of reactor,  $Q_0$  – flow rate of wastewater entering into the system.

For a system to run efficiently, the theoretical and experimental MHRT should be equal. However, this is seldom true for most industrial systems. The dead zone in the system can be calculated by Eq. (2):

$$(2) \quad V_d = \left(1 - \frac{\bar{t}}{\tau}\right)$$

where,  $V_d$  – fraction of dead volume,  $\bar{t}$  – experimental MHRT and  $\tau$  – theoretical MHRT.

Different models (tank-in-series, axial dispersion, tank-in-series with backmixing and tank-in-series with dead volumes, etc.) were applied to model the experimental RTD data to obtain a physical representation of the hydrodynamics inside the aeration tank and clarifier. The RTD software DTSPRO V. 4.21 developed by PROGEPI [25, 26] was used to model the measured RTD data in the aeration tank and secondary clarifier.

#### Analysis and modelling of the imperfect input impulse using convolution

In industry, the systems are interconnected and processed material leaving one reactor serves as the input to the succeeding one (see Fig. 2). These processes may have different hydrodynamic characteristics, and the RTD study of each individual

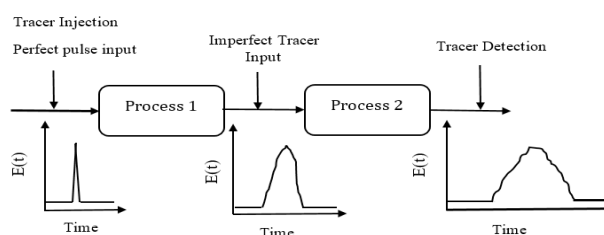


Fig. 2. Interference with the input signal due to the non-ideal impulse input.

system can be very lengthy and tiresome. This problem can be evaluated by convolution integrals [27]. Mathematically, convolution can be defined as the operation between two functions or signals that produces a third function which is correlated to the original function. The radiotracer that enters a process from the previous reactor serves as an imperfect input impulse and the output signal gets modified. This signal modification is related to the  $E$  curve of this vessel according to the convolution integral in Eq. (3):

$$(3) \quad C_{out}(t) = \int_0^t C_{in}(t-t')E(t')dt'$$

where  $C_{out}$  – output tracer concentration,  $C_{in}$  – input tracer concentration and  $E(t)$  – normalized RTD at the outlet of the system.

This equation can be used to obtain a non-distorted  $E$  curve for the system when the input and output RTD signals of the system are known.

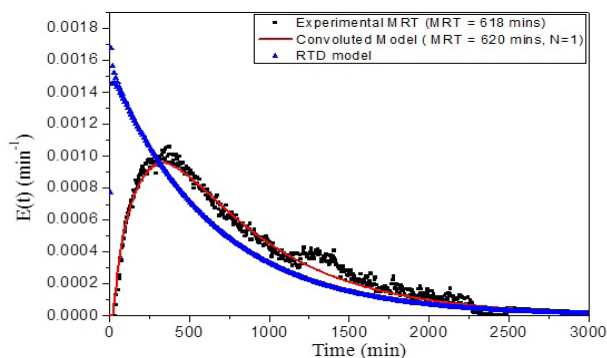
RTD models are physical representations of flow in the reactor. The RTD data obtained experimentally can be conveniently modelled to study the deviation of real reactors from ideal behaviour. However, systems with imperfect impulse inputs cannot be directly modelled using RTD models present in the literature. In such a case, a suitable RTD model is selected and the model  $E(t)$  is obtained by assuming its parameter values. The model  $E(t)$  is then convoluted with the input signal of the system to obtain a convoluted model that fits to the corresponding experimental RTD data.

### Results and discussion

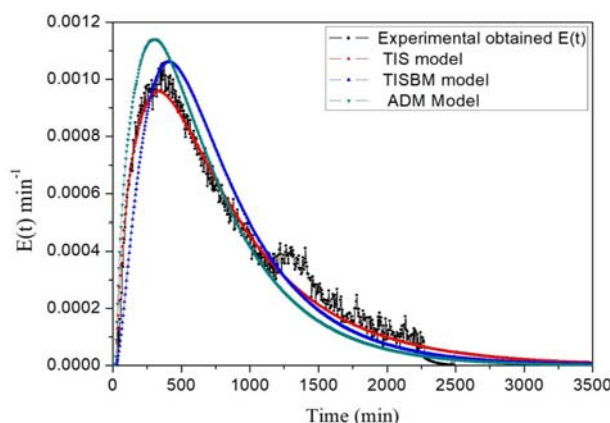
#### Aeration tank

The second experiment performed on the two-stage ASP was monitored at the outlet of aeration tank 2 (D9) as shown in Fig. 1. The RTD curve obtained at the outlet of aeration tank 2 was pre-corrected to remove any signal disturbances. As can be perceived from Fig. 1, no equalization tank exists prior to the aeration tank, hence, a temporary variation of flow rate in the system may have caused the sudden disturbance in signal observed during a later stage of the experiment. The RTD curve did not display any multiple peaks which also indicates that no specific bypassing and parallel flow paths exist in the tank. The volume of the aeration tank was 3240 m<sup>3</sup> and the volumetric flow rate entering the aeration tank during the experiment was

$5.21 \pm 1\%$   $\text{m}^3/\text{min}$ . Therefore, the theoretical MHRT was estimated to be 622 min, as reported in Table 1. The non-ideal impulse response, i.e. the RTD curve measured at the outlet of the aeration tank, was normalized to obtain  $E(t)$  curves. As ideally the aeration tank is supposed to act as a perfect continuously stirred tank reactor (CSTR), the tank-in-series (TIS) model was chosen to simulate the tank. The model normalized the RTD, and the  $E_m(t)$  was obtained from the model equation and convoluted against the non-ideal input signal of the aeration tank (D6). The model parameter, i.e. number of tanks (N), was varied to acquire the curve that fits the experimental  $E(t)$  curve best. The reactor was best represented with a single mixing tank ( $N = 1$ ), implying that the aeration tank acted as an ideal mixing tank with negligible anomalies. The model MHRT was calculated from the model parameters and was found to be 620 min. A comparison of the experimental and convoluted models is shown in Fig. 3. As the theoretical and experimentally measured MHRTs are approximately equal, no dead volume is present within the aeration tank. This implies that



**Fig. 3.** RTD curve monitored at the outlet of aeration tank (D9) and model adjustment by convolution.



**Fig. 4.** Comparison of RTD models for the aeration tank.

almost the entire geometric volume of the tank is available for the aeration process. Tank-in-series and tank-in-series with backmixing are the most suitable models for representing aeration tanks [28, 29]. Systems with surface aerators or baffles often exhibited significant backmixing in the tank. Often complex aeration systems are also represented by compartment models that consist of a combination of continuously stirred tank reactor (CSTR) and plug flow reactor (PFR) to account for dead zones, bypassing and recycle lines [25]. Three different RTD models that are detailed in the literature (i.e. TIS, TISBM and ADM) were simulated for the aeration tank as shown in Fig. 4 and the model parameters are reported in Table 2.

### Secondary clarifier

The aeration tank and secondary clarifier were connected in series, the radiotracer concentration at the outlet of the aeration tank (D9) served as an input signal for the clarifier. Since the volume of the secondary clarifier was  $890 \text{ m}^3$  and the total volumetric flow rate entering the secondary clarifier was  $5.21 \text{ m}^3/\text{min}$ , the theoretical mean residence time (MRT) was determined to be 174 min. The secondary clarifier should ideally act as a perfect plug flow reactor, however, deviation from the same can be suitably displayed by the axial dispersion model. The axial dispersion model was used to simulate the normalized RTD signal at the outlet of the secondary clarifier (D10) and the model  $E_m(t)$  was convoluted with the input radiotracer signal of the secondary clarifier (D9). The obtained Peclet number (Pe) for the axial dispersion component was 10. This high value of Pe indicates that the clarifier acts as a plug flow reactor. The model MRT was calculated from the model parameters and was found to be 66 min. The comparison of the theoretical and experimental MHRTs predicts that a proportion of the reactor volume is inactive and unavailable for hydraulic flow. The dead zone in the reactor can be explained due to the accumulation of biomass at the bottom of the tank. Figure 5 shows the comparison of the experimental and theoretical model simulated curves corresponding to the optimum model parameters. The comparison of the TIS, TISBM and ADM models for the secondary clarifier is shown in Fig. 6 and the model parameters reported in Table 2.

### Conclusion

The hydraulic behaviour of the aeration tank and secondary clarifier was successfully studied at an

**Table 2.** Comparison of different models and their parameters for the aeration tank and secondary clarifier

RTD models	Aeration tank			Secondary clarifier		
	MRT [min]	Parameters	R <sup>2</sup>	MRT [min]	Parameters	R <sup>2</sup>
Tank-in-series model (TIS)	620	N = 1	0.99	71	N = 6	0.93
Tank-in-series with backmixing model (TISBM)	600	N = 2, $\alpha = 1$	0.97	72	N = 6, $\alpha = 0.4$	0.97
Axial dispersion model (ADM)	610	Pe = 0.1	0.98	70	Pe = 10	0.99

Pe – Peclet number. N – number of the tank in the tank-in-series model.  $\alpha$  – backmixing ratio.

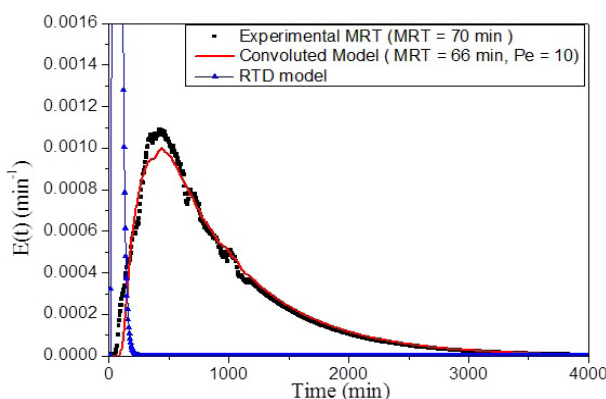


Fig. 5. RTD curve monitored at the outlet of secondary clarifier (D10) and model adjustment by convolution.

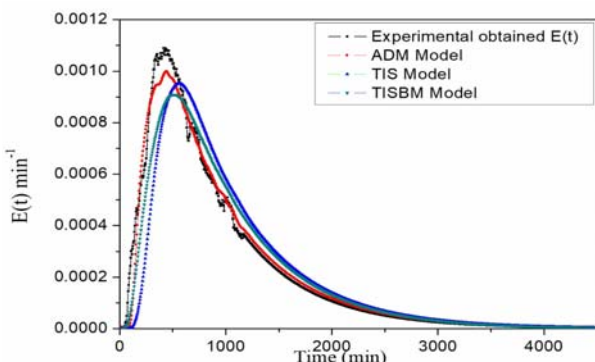


Fig. 6. Comparison of RTD models for the secondary clarifier.

industrial scale effluent treatment plant of a paper manufacturer in India. The aeration tank was found to work very efficiently with no dead zones or bypassing stream present in the tank. The convolution method was applied along with the suitable RTD model to obtain convoluted models that fit the experimentally obtained RTD data well. The hydrodynamic model for the aeration tank was suitably represented with a single mixing tank further validating that the aeration tank works as a perfect mixing tank. A simple axial dispersion model was found to be suitable to describe the flow behaviour of the secondary clarifier.

**Acknowledgment.** The authors are grateful to the Board of Research in Nuclear Sciences (BRNS) and the Department of Atomic Energy (DAE) in Mumbai, India for funding the work under the project (35/14/09/2015-BRNS/3069) and to Shreyans Industries Ltd. for providing the necessary support to conduct the study at their effluent treatment plant.

## References

1. Bajpai, P. (2001). Microbial degradation of pollutants in pulp mill effluents. *Adv. Appl. Microbiol.*, 48, 79–134. DOI: 10.1016/S0065-2164(01)48001-4.
2. Pokhrel, D., & Viraraghavan, T. (2004). Treatment of pulp and paper mill wastewater – a review. *Sci. Total Environ.*, 333(1/3), 37–58. DOI: 10.1016/j.scitotenv.2004.05.017

3. Thompson, G., Swain, J., Kay, M., & Forster, C. (2001). The treatment of pulp and paper mill effluent: a review. *Bioresour. Technol.*, 77(3), 275–286.
4. Phiri, O., Mumba, P., Moyo, B. H. Z., & Kadewa, W. (2005). Assessment of the impact of industrial effluents on water quality of receiving rivers in urban areas of Malawi. *Int. J. Environ. Sci. Technol.*, 2(3), 237–244. DOI: 10.1007/bf03325882.
5. Suthar, S., Nema, A. K., Chabukdhara, M., & Gupta, S. K. (2009). Assessment of metals in water and sediments of Hindon River, India: impact of industrial and urban discharges. *J. Hazard. Mater.*, 171(1), 1088–1095. DOI: 10.1016/j.jhazmat.2009.06.109.
6. Metcalf & Eddy Inc., Burton, F. L., Stensel, H. D., & Tchobanoglous, G. (2003). *Wastewater engineering: Treatment and reuse* (4th ed). New Delhi: McGraw-Hill.
7. Peavey, H. S., Rowe, D. R., & Tchobanoglous, G. (2013). *Environmental engineering* (3rd ed.). New Delhi: McGraw-Hill.
8. International Atomic Energy Agency. (2008). *Radiotracer residence time distribution method for industrial and environmental applications*. Vienna: IAEA. (Training Course Series No. 31). Available from <http://www-pub.iaea.org/books/IAEABooks/7891/Radiotracer-Residence-Time-Distribution-Method-for-Industrial-and-Environmental-Applications>.
9. International Atomic Energy Agency. (2011). *Radiotracer applications in wastewater treatment plants*. Vienna: IAEA. (Training Course Series No. 49). Available from <http://www-pub.iaea.org/books/IAEABooks/8552/Radiotracer-Applications-in-Wastewater-Treatment-Plants>.
10. Koennecke, H. G., Luther, D., Koepping, K., Schoen, J., & Ulrich, H. (1984). Studies of the hydrodynamic properties in biological clarification plants using radiotracers. *Clean-Soil, Air, Water*, 12(4), 431–434. DOI: 10.1002/aheh.19840120416.
11. Othman, N., & Kamarudin, S. K. (2014). Radiotracer technology in mixing processes for industrial applications. *Sci. World J.*, 2014, 768604. DOI: 10.1155/2014/768604.
12. Pant, H., Kundu, A., & Nigam, K. (2001). Radiotracer applications in chemical process industry. *Rev. Chem. Eng.*, 17(3), 165–252. DOI: 10.1515/REVCE.2001.17.3.165.
13. Pant, H. J., Sharma, V. K., Singh, G., Raman, V. K., Bornare, J., & Sonde, R. R. (2012). Radiotracer investigation in a rotary fluidized bioreactor. *J. Radioanal. Nucl. Chem.*, 294(1), 59–63. DOI: 10.1007/s10967-011-1511-2.
14. Audic, J., Fayoux, C., Lesty, Y., & Brisset, P. (1994). Sludge retention times in clarifier: a key point for population dynamic and nutrients removal control. *Water Sci. Technol.*, 29(7), 57–60.
15. Battaglia, A., Fox, P., & Pohland, F. (1993). Calculation of residence time distribution from tracer recycle experiments. *Water Res.*, 27(2), 337–341. DOI: 10.1016/0043-1354(93)90093-W.
16. Borroto, J., Dominguez, J., Griffith, J., Fick, M., & Leclerc, J. (2003). Technetium-99m as a tracer for the liquid RTD measurement in opaque anaerobic digester: application in a sugar wastewater treatment plant. *Chem. Eng. Process-Process Intensification*, 42(11), 857–865.
17. Chmielewski, A. G., Owczarczyk, A., & Palige, J. (1998). Radiotracer investigations of industrial waste water equalizer-clarifiers. *Nukleonika*, 43(2), 185–194.
18. Kasban, H., Zahran, O., Arafa, H., El-Kordy, M., Elaraby, S. M., & El-Samie, F. A. (2010). Laboratory

- experiments and modeling for industrial radiotracer applications. *Appl. Radiat. Isot.*, 68(6), 1049–1056. DOI: 10.1016/j.apradiso.2010.01.044.
19. Sánchez, F., Viedma, A., & Kaiser, A. (2016). Hydraulic characterization of an activated sludge reactor with recycling system by tracer experiment and analytical models. *Water Res.*, 101, 382–392. DOI: 10.1016/j.watres.2016.05.094.
  20. Levenspiel, O. (2001). *Chemical reaction engineering* (3rd ed.). New Delhi: Wiley.
  21. Fogler, S. H. (2011). *Elements of chemical reaction engineering* (4th ed.). New Delhi: PHI.
  22. Sarkar, M., Sangal, V. K., Sharma, V. K., Samantray, J., Bhunia, H., Bajpai, P. K., Kumar, A., Naithani, A. K., & Pant, H. J. (2017). Radiotracer investigation and modeling of an activated sludge system in a pulp and paper industry. *Appl. Radiat. Isot.*, 130, 270–275. DOI: 10.1016/j.apradiso.2017.10.016.
  23. Kim, H. S., Shin, M. S., Jang, D. S., & Jung, S. H. (2006). Indepth diagnosis of a secondary clarifier by the application of radiotracer technique and numerical modeling. *Water Sci. Technol.*, 54(8), 83–92. DOI: 10.2166/wst.2006.857.
  24. Othman, N., Hassan, N. P. I. M., Yahya, R., Adnan, M. A. K., Shari, M. R., Hassan, H., & Mahmood, A. A. (2017). Radiotracer application in Malaysia: Residence Time Distribution study. *Malays. J. Anal. Sci.*, 21(2), 445–451. DOI: 10.17576/mjas-2017-2102-20.
  25. Farooq, M., Khan, I., Gul, S., Palige, J., & Dobrowolski, A. (2003). Radiotracer investigations of municipal sewage treatment stations. *Nukleonika*, 48(1), 57–61.
  26. Leclerc, J., Schweich, D., Bernard, A., & Detrez, C. (1995). DTS: a Software Package for Flow Simulation in Reactors. *Oil & Gas Science and Technology-Rev. IFP*, 50(5), 641–656. DOI: 10.2516/ogst:1995039.
  27. Gutierrez, C. G., Dias, E. F., & Gut, J. A. (2010). Residence time distribution in holding tubes using generalized convection model and numerical convolution for non-ideal tracer detection. *J. Food Eng.*, 98(2), 248–256. DOI: 10.1016/j.jfoodeng.2010.01.004.
  28. Lemoulllec, Y., Potier, O., Gentric, C., & Leclerc, J. -P. (2008). A general correlation to predict axial dispersion coefficients in aerated channel reactors. *Water Res.*, 42(6), 1767–1777. DOI: 10.1016/j.watres.2007.10.041.
  29. Chmielewski, A. G., & Selecki, A. (1979). Application of radioactive tracers to some processes on a large scale. *Isotopenpraxis-Isot. Environ. Health Stud.*, 15(9), 276–281.