



Internals Design of Continuous Stirred Tank Electrochemical Reactor Based on the Residence Time Distribution Approach

Raghad S. Mahmood¹ Ammar S. Abbas^{2*}

Chemical Engineering Department, College of Engineering, University of Baghdad



CrossMark

Abstract

The design of the reactor internals of the two-phase Continuous Stirred Tank Electrochemical Reactor was studied under different conditions. Two types of air distributors were tested (cubic and circular) according to the bubble size produced by each, measured by a high-speed camera. Two impeller types were tested (Rushton turbine 4 and 6 blades). The results indicate that the bubble size increases with increasing airflow, but the cubic distributor has a smaller bubble diameter than the circular distributor under all conditions. The value of average time for the 4-blades turbine was more than it for the 6-blades turbine at a lower mixing speed, but at 600 rpm, the 6-blade turbine has a value of average time closer to the ideal space-time.

Keywords: Electrochemical, advanced oxidation, reactor, hydrodynamics, bubble size, mixer

1. Introduction

Many industrial processes generate toxic and polluted wastewaters that are difficult to degrade and require costly physical or physiochemical treatments [1]. Traditional treatment methods have become inadequate because of their limitations when used individually [2]. Various techniques have been applied, such as electro-advanced oxidation technologies (EAOP). EAOP is distinguished by its capacity to make use of the high reactivity of hydroxyl radicals, which are produced from chemical precursors such as light irradiation [3].

Pollutants can be removed during electrochemical treatment by either direct anodic oxidation [4], [5], or indirect oxidation [11]. The electro-Fenton technique has sparked a lot of attention among indirect EAOPs for wastewater remediation. In the electro-Fenton method, oxygen is necessary to generate hydrogen peroxide (H_2O_2). H_2O_2 decomposes catalytically to produce the strong $\bullet OH$ radicals that mediate the degradation of organic contaminants [6].

In ideal reactors like the Continuous Stirred Tank Reactor (CSTR), perfect mixing is assumed to study reactor performance [7]. However, most fluid flow through the chemical reactors used in industry is not ideal [8].

Understanding the non-ideal behavior is essential when designing a reactor. Reactor designers aim to get real reactors to behave as closely as possible to the ideal reactors [9]. In addition, hydrodynamics, fluid flow behavior, and mixing conditions are critical

parameters for meeting system requirements and overcoming troubles that typically result in reduced reactor performance [10]. Non-ideal behavior is caused by a variety of factors, including fluid channeling as it moves through the reaction vessel, longitudinal mixing caused by vortices and turbulence, the presence of stagnant regions inside the reactor, bypassing or short-circuiting, impeller, or other mixing device failure to supply perfect mixing and so on [11], [12]. Studying the flow behavior and mixing conditions through reactors using residence time distribution (RTD) studies has been one of the most critical aspects of understanding vessel hydrodynamics. RTD is a property of mixing in the chemical reactor that determines how long the reaction mixture has been in the reactor [13], [14].

The bubbles behavior in gas-liquid stirred tanks is critical, particularly in supplying oxygen from the gas phase to the liquid phase, which is required for the electro-Fenton oxidation to generate $\bullet OH$ radicals [15]. One of the most critical parameters impacting the efficiency of this form of operation is the contact time of air bubbles with the liquid. The contact time is dependent on the size of the bubble, its terminal velocity, diffuser submergence, and water velocity [R]. The bubble size distribution is a significant parameter in a bubble reactor and has a great impact on the performance of gas-liquid contactors [16], [17] thus, understanding the flow dynamics and mass transfer processes between phases [18], [19]. Bubbles rising in a highly turbulent fluid are likely to have a

*Corresponding author e-mail: ammaraabbas@coeng.uobaghdad.edu.iq

Receive Date: 20 February 2022, Revise Date: 20 April 2022, Accept Date: 21 April 2022.

DOI: [10.21608/ejchem.2022.122976.5502](https://doi.org/10.21608/ejchem.2022.122976.5502).

©2023 National Information and Documentation Center (NIDOC).

more complex behavior [20].

Mixing and agitation of liquids in stirred tanks are used by many industries, including chemical, biotechnological, pharmaceutical, and food processing, to increase heat and mass transfer rates, prevent particle settling, obtain emulsions, and unification all physical property gradients [21]. The mixing processes in an agitated tank are influenced by hydrodynamics. As a result, evaluating the level of mixing and the overall behavior and performance of agitated tanks and analyzing agitation hydrodynamics are crucial for process quality and economics [22].

Because of the complicated nature of impeller-induced turbulence in agitated vessels, inconsistencies in mixing quality might be linked to a lack of clear understanding of the mixing processes [23]. In order to determine mixing efficiency and carefully anticipate the total performance of these systems, in-depth research is required. The performance of mixing processes has been proven to be a function of mixing time, impeller type, number of impeller blades, blade size, working angular speeds, and vessel configurations [24].

The work aims to characterize the best design for the electrochemical reactor with two-phase flow based on the choice of the best air distributor via the bubble size distribution, and the best mixer using RTD approach.

2. Experimental work

2.1. Bubble size measurement

In order to choose the best air diffuser, the dimensions of the bubbles were photographed using a camera (Nikon D750, 1/20000s); images were captured near the reactor's wall nearest to the camera, which was pointing at the reactor's centerline, to reduce image distortion. The bubbles along the wall are assumed to be typical of those within the reactor in this situation. Two types of distributors were tested, cubic and circular, as shown in Figure (1). Each air distributor was photographed at 0.3, 0.5, and 1 L/min airflow rate then the images were processed by program ImageJ [25] to calculate the size of bubbles.

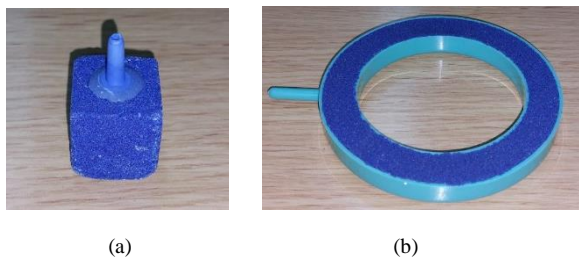


Fig. 1. Type of air distributor (a: circular; b: cubic)

2.2. Reactor setup

Two types of Rushton turbine (4 and 6 blades) shown in Figure 2 were tested as a mixer. The mixer is installed at the bottom of the rotating electrode (1.5 cm in diameter and 6 cm in length) to preserve its surface area. RTD was studied with these impellers at 100 ml/min water flow rate (as test fluid) and 300, 450, 600 rpm. The diameter of the reactor was 15 cm, its length was 24 cm (filled up to 80%), and it had 4 baffles with a width of 1.5 cm. The pulse response technique was used to measure the RTD of the liquid age distribution using methylene blue as a tracer. At time zero, 30 mL of the tracer (500 ppm concentration) was injected, and the computer in-line photometer measured the resistance of the outflow stream. The outlet concentration versus time ($C(t)$) can be measured.

3. Results and discussion

3.1. Bubble size measurement

The photographic method is used to determine the size of the bubbles at a different air flow rate (0.3; 0.5; 1 L/min).

Figures (3) and (4) show the photographed bubbles at 0.3; 0.5; 1 L/min for cubic and circular distributors, respectively. The only clear images in all photographs are of bubbles near the vessel wall. Some bubbles do not have a perfect sphere shape, but an oblate spheroid can be used to approximate it. It can also be observed that the bubble size is not uniform in all images. The imageJ program processed the images, and the average diameters of the bubble size for each distributor were calculated.

Table (1) represents the average diameter value for the bubbles of cubic and circular distributors. At 0.3 L/min, the size of the bubbles for the cubic distributor was equal to 1.1692 mm, which is smaller than the size of the bubbles for the circular distributor (1.4248 mm). As the airflow increases, the bubble size of both distributors increases from 1.1692 mm to 1.3159 mm for the cubic distributor and from 1.4248 mm to 1.8083mm for the circular distributor, but the bubble size of the cubic distributor remains smaller than the bubble size of the circular distributor.



(a)

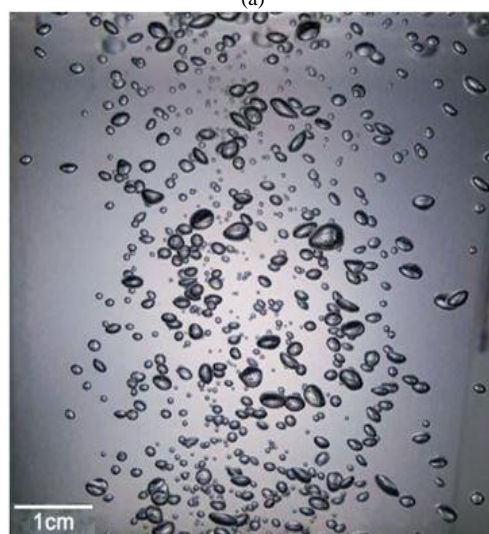


(b)

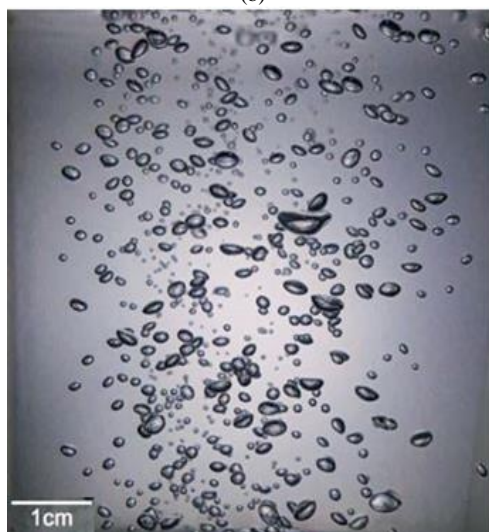
Fig. 2. Type of air distributor (a: circular; b: cubic)



(a)

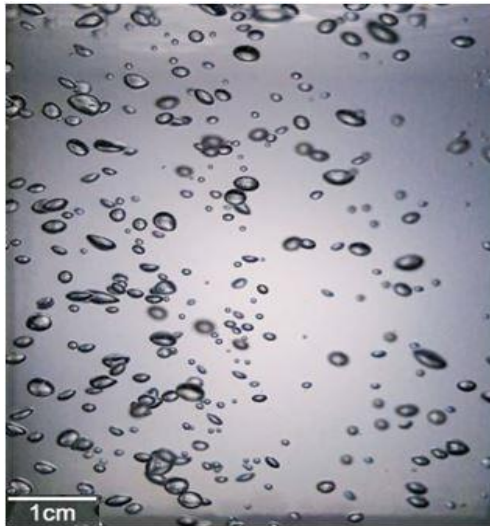


(b)

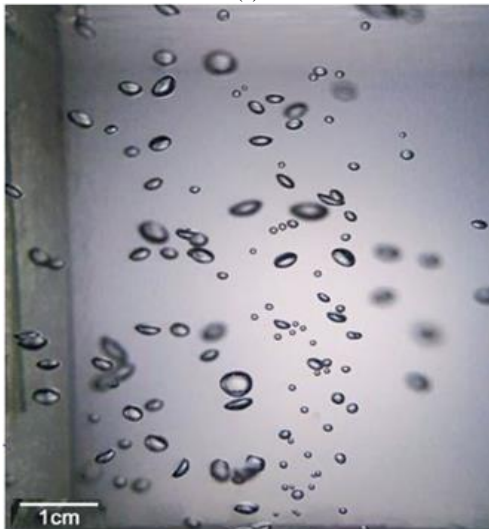


(c)

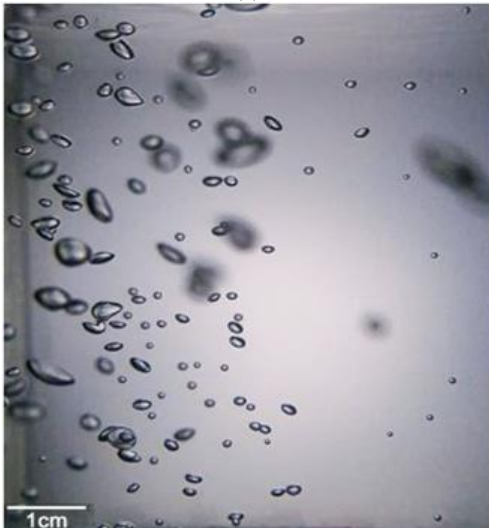
Fig. 3. Cubic distributor bubbles (a: 0.3L/min; b: 0.5 L/min; c: 1L/min)



(a)



(b)



(c)

Fig. 4. Cubic distributor bubbles (a: 0.3L/min; b: 0.5 L/min; c: 1L/min)

This increase in the size of the bubbles occurred because the airflow rate directly contributes to the bubble expansion process during bubble formation. The higher the flow rate, the faster the bubble formed, resulting in a larger bubble size. Since it is known that the smallest bubble size is the best air distributor thus, the cubic distributor has been selected for the exit age experiments.

Table 1
The value of average diameter for the bubbles of cubic and circular distributor

Airflow rate, L/min	Average diameter (mm)	
	Cubic distributor	Circular distributor
0.3	1.1692	1.4248
0.5	1.2983	1.7677
1	1.3159	1.8083

3.2. Choice the best mixer

The experimental exit age distribution (RTD) can be determined by using Eq. (1)

$$E_{(t)} = \frac{c_{(t)}}{\int_0^{\infty} c_{(t)} dt} \quad (1)$$

The average time is given by Eq. (2).

$$\bar{t} = \int_0^{\infty} tE_{(t)} dt \quad (2)$$

If the reactor is free of dysfunctions (dead volume, short circuit), the average time will equal the ideal space-time, which is calculated by Eq. (3).

$$\tau = \frac{V}{Q} \quad (3)$$

Figure (5) represent the experimental exit age distribution value versus time at different mixing speed (a: four- blades turbine; b: six-blade turbine).

As expected for a perfect CSTR, all of these curves exhibit a rapid increase at the beginning, followed by an exponential fall. In addition, as shown in Figure 5 (a) that the highest peak (less residence time distribution) at 300 rpm and the lowest peak (higher residence time distribution) at 600 rpm. But the results are almost equal in the case of the six-blade turbine at all mixing speeds (Figure 5 (b)).

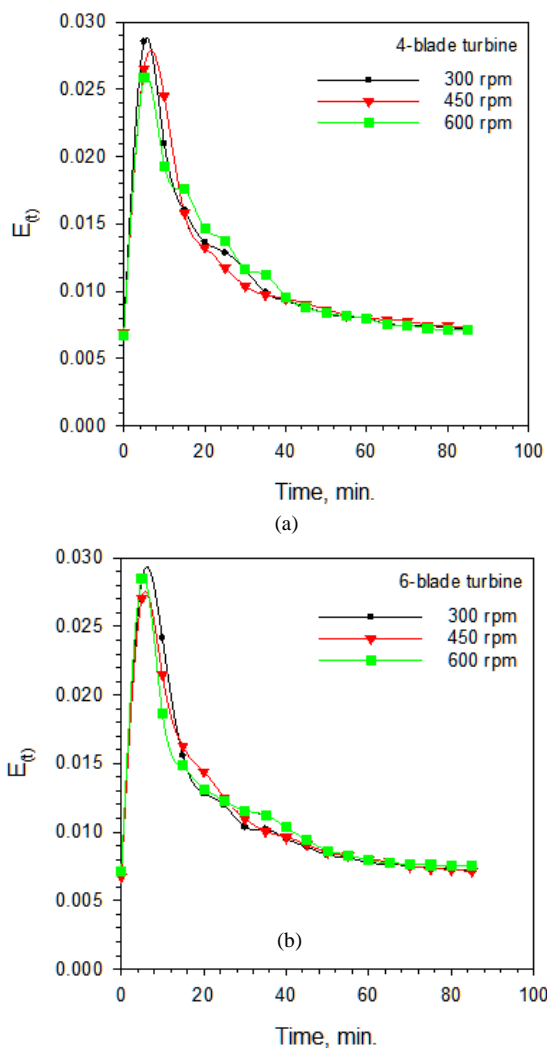


Fig. 5. The experimental exit age distribution value versus time at different mixing speeds (a: four-blade turbine; b: six-blade turbine)

Table 2 shows the value of the average experimental time for both, four-blade turbine and the six-blade turbine. It can be seen from the table that when the mixing speed increases, the average time increases from 33.83 min to 34.06 min for the four-blade turbine and from 33.63 min to 34.58 min for the six-blade turbine. The value of average time increased with increased mixing speed because in a bubbling reactor, mixing promotes gas bubble rotation, which increases the flow route of gas bubbles in the bubbling reactor. As a result, the gas bubble's residence time increases, and the chemical reaction between the gas bubble and mixed liquid oxidants are more fully completed, increasing the efficiency of the electrochemical reactor [26].

Table 2: The value of average experimental time for four and six-blade turbine

Mixing speed, rpm	Ideal space-time, min.	average time using four-blade turbine, min.	Average time six-blades turbine, min.
300	34.64	33.83	33.63
450		34.02	33.99
600		34.06	34.58

In addition, it can be observed from Table 2 that the value of the average time of the four-blade turbine at 300 rpm (33.83 min) is more than that of the six-blade turbine (33.63 min). At 450 rpm the value of the average time of the four-blade turbine (34.02 min) is also more than that of the six-blade turbine (33.99 min) but it still less than the ideal space-time (34.64 min). At 600 rpm the results were reversed where the six-blade turbine has a higher average time value than the four-blade turbine, which is equal to 34.58 min and 34.04 min, respectively.

Although the average time value for the four-blade turbine is more than the average time for the six-blade turbine at 300 rpm and 450 rpm, the six-blade turbine was chosen because at 600 rpm it gives an average time value equal to 34.58 min which is closer to the ideal space-time value (34.64 min).

4. Conclusions

Various tests were carried out to evaluate hydrodynamics and mixing behavior in the continuous stirred tank electrochemical reactor under various operating conditions (type of distributor, type of impeller, mixing speed, and airflow rate), as well as to identify the best set of variables.

Two types of distributors were tested, cubic and circular, by photographing the bubbles at 0.3; 0.5; 1 L/min by using high-speed camera. The results indicate that the average diameter for the cubic distributor was less than it for the circular distributor, where it was 1.1692 mm for cubic distributor at 0.3 L/min but for circular distributor equal to 1.4248 mm at the same airflow. When the airflow increases, the bubble size increases to 1.3159 mm for the cube distributor, but still smaller than the bubble size for the circular distributor (1.8083 mm)

Two types of the mixer were tested (Rushton turbine) 4, 6 blades. The results show that the average time for the 4-blades impeller was more than it for the 6-blades impeller at 300 and 450 rpm. But at 600 rpm this result was reversed where the average time for the 4-blades impeller was less than the 6-blades impeller. However, the four-blade mixer was chosen for the remainder experiments because it gives the value of the average time close to the ideal one at a lower mixing speed.

References

- [1] B. K. Körbahti and A. Tanyolaç, "Modeling of a continuous electrochemical tubular reactor for phenol removal," *Chem. Eng. Commun.*, vol. 190, no. 5–8, pp. 749–762, 2003, doi: 10.1080/00986440302129.
- [2] J. Sendhil, P. K. A. Muniswaran, and C. Ahmed Basha, "Residence time distribution studies in flow through tubular electrochemical reactor," *Int. J. Eng. Res. Dev.*, vol. 1, no. 7, pp. 52–62, 2012, [Online]. Available: www.ijerd.com.
- [3] I. Tröster et al., "Electrochemical advanced oxidation process for water treatment using DiaChem® electrodes," *Diam. Relat. Mater.*, vol. 11, no. 3–6, pp. 640–645, 2002, doi: 10.1016/S0925-9635(01)00706-3.
- [4] C. A. Martínez-Huitle and S. Ferro, "Electrochemical oxidation of organic pollutants for the wastewater treatment: Direct and indirect processes," *Chem. Soc. Rev.*, vol. 35, no. 12, pp. 1324–1340, 2006, doi: 10.1039/b517632h.
- [5] M. Panizza and G. Cerisola, "Direct And Mediated Anodic Oxidation of Organic Pollutants," *Chem. Rev.*, vol. 109, pp. 6541–6569, 2009.
- [6] Z. I. Abbas and A. S. Abbas, "Oxidative degradation of phenolic wastewater by electro-fenton process using MnO₂-graphite electrode," *J. Environ. Chem. Eng.*, vol. 7, no. 3, p. 103108, 2019, doi: 10.1016/j.jece.2019.103108.
- [7] A. H. Khamthani, "Statistical Studies of a Real Continuous Stirred Tank Reactor (CSTR) Based on Experimental Data," *Universiti Teknologi PETRONAS*, 2005.
- [8] O. Levenspiel, *Chemical reaction engineering*, 3rd ed. New York: John Wiley & Sons, 1999.
- [9] M. E. Obonukut and P. G. Basseyy, "Residence Time Distribution of A Tubular Reactor," *Int. J. Sci. Res. Educ.*, vol. 4, no. 1, pp. 4767–4777, 2016, doi: 10.18535/ijre/v4i01.02.
- [10] D. Olivet, J. Valls, M. À. Gordillo, À. Freixó, and A. Sánchez, "Application of residence time distribution technique to the study of the hydrodynamic behaviour of a full-scale wastewater treatment plant plug-flow bioreactor," *J. Chem. Technol. Biotechnol.*, vol. 80, no. 4, pp. 425–432, 2005, doi: 10.1002/jctb.1201.
- [11] D. Rajavathsavai, "Study of Hydrodynamic and Mixing Behaviour of Continuous Stirred Tank Reactor Using CFD Tools," *National Institute of Technology*, January, 2012.
- [12] H. Scott Fogler, *Elements of chemical reaction engineering*, 3rd ed., vol. 42, no. 10. Asoke K. Ghosh, 1999.
- [13] A. Khapre, D. Rajavathsavai, and B. Munshi, "Study on residence time distribution of CSTR using CFD," *Indian J. Chem. Technol.*, vol. 23, no. 2, pp. 114–120, 2016.
- [14] R. S. Mahmood and A. S. Abbas, "Validation of a Three-parameters Hydrodynamic Model to Describe the non-ideal Flow in a Continuous Stirred Tank Reactor of the Electro-Fenton Oxidation of Organic Pollutants in Wastewater," *J. Phys. Conf. Ser.*, vol. 1973, no. 1, p. 012092, 2021, doi: 10.1088/1742-6596/1973/1/012092.
- [15] H. Wang, X. Jia, X. Wang, Z. Zhou, J. Wen, and J. Zhang, "CFD modeling of hydrodynamic characteristics of a gas-liquid two-phase stirred tank," *Appl. Math. Model.*, vol. 38, no. 1, pp. 63–92, 2014, doi: 10.1016/j.apm.2013.05.032.
- [16] W. H. Zhang, X. Jiang, and Y. M. Liu, "A method for recognizing overlapping elliptical bubbles in bubble image," *Pattern Recognit. Lett.*, vol. 33, no. 12, pp. 1543–1548, 2012, doi: 10.1016/j.patrec.2012.03.027.
- [17] D. S. Mavinic and J. K. Bewtra, "Bubble size and contact time in diffused aeration systems," *J. Water Pollut. Control Fed.*, vol. 46, no. 9, pp. 2129–2137, 1974.
- [18] M. Senouci-Bereksi, F. K. Kies, and F. Bentahar, "Hydrodynamics and Bubble Size Distribution in a Stirred Reactor," *Arab. J. Sci. Eng.*, vol. 43, no. 11, pp. 5905–5917, 2018, doi: 10.1007/s13369-018-3071-z.
- [19] R. Bi et al., "Experimental Study on Bubble Size Distribution in Gas-Liquid Reversed Jet Loop Reactor," *Int. J. Chem. React. Eng.*, vol. 18, no. 1, pp. 1–14, 2020, doi: 10.1515/ijcre-2019-0102.
- [20] M. S. N. Oliveira, A. W. Fitch, and X. Ni, "A STUDY OF BUBBLE VELOCITY AND BUBBLE RESIDENCE TIME IN A GASSED OSCILLATORY BAFFLED COLUMN Effect of Oscillation Frequency," *Engineering*, vol. 81, no. 2, 2003.
- [21] H. M. Issa, "Retracted: Power consumption, mixing time, and oxygen mass transfer in a gas-liquid contactor stirred with a dual impeller for different spacing," *J. Eng. (United Kingdom)*, vol. 2016, 2016, doi: 10.1155/2017/3906268.
- [22] K. Yapici, B. Karasozen, M. Schäfer, and Y. Uludag, "Numerical investigation of the effect of the Rushton type turbine design factors on agitated tank flow characteristics," *Chem. Eng. Process. Process Intensif.*, vol. 47, no. 8, pp. 1340–1349, 2008, doi: 10.1016/j.cep.2007.05.002.
- [23] F. Delvigne, J. Destain, and P. Thonart, "Structured mixing model for stirred bioreactors: An extension to the stochastic approach," *Chemical Engineering Journal*, vol. 113, pp. 1–12, 2005, doi: 10.1016/j.cej.2005.06.007.
- [24] I. Torotwa, C. Ji, "A Study of the Mixing Performance of Different Impeller Designs in Stirred Vessels Using Computational Fluid

- Dynamics, *Des.* 2018, Vol. 2, **2**, 2018, <https://doi.org/10.3390/DESIGNS2010010>.
- [25] C. A. Schneider, W. S. Rasband, and K. W. Eliceiri, "NIH Image to ImageJ: 25 years of image analysis," *Nat. Methods*, vol. 9, no. 7, pp. 671–675, 2012, doi: 10.1038/nmeth.2089.
- [26] D. Zhang, H. Tao, C. Yao, and Z. Sun, "Effects of residence time on the efficiency of desulfurization and denitrification in the bubbling reactor," *Chem. Eng. Sci.*, vol. 174, pp. 203–221, 2017, doi: 10.1016/j.ces.2017.09.010.