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# **ORIGINAL ARTICLE**

# Hydrodynamic study of a novel palm shell Granular activated carbon twin-chamber upflow Bio-electrochemical reactor for Sequential Nitrification and Denitrification

# Seyyed Alireza Mousavi<sup>1</sup>, Shaliza Ibrahim<sup>2</sup>

<sup>1</sup>Department of Environmental Health Engineering, Kermanshah University of Medical Sciences, Kermanshah, Iran.

<sup>2</sup>Department of Civil Engineering, Faculty of Engineering, University of Malaya, 50603 Kuala Lumpur, Malaysia.

Email: seyyedarm@yahoo.com

# **ABSTRACT**

The hydrodynamic characteristics of palm shell granular activated carbon twin-chamber upflow bio-electrochemical reactor (PSGAC-TCUBR) were studied at different values of HRT, and current intensity in the range of 6-24h, and 50 mA, respectively. The tracers test of PSGAC-TCUBR (under abiotic and bioelectrochemical conditions) was studiedvia investigation of residence time distribution (RTD), according to an appropriate method. First, reactors run without applying electric current to determine and to develop the analytical RTD function in both compartments. In the next stage of the experiments, a reactor was run as a bio-electrochemical reactor, and the RTD profiles described quantitatively how long a portion of the fluid has spent in the reactor by measuring the concentration of tracers in effluent. The results for different flow rates at different type of tracers show curves with different patterns, a peak at the beginning and a long tail that is distorted by the solution flow rate. The results for different flow rates show curves with similar patterns. At low flow rates, both the dispersion and the interaction with a stagnant zone arestrong. The hydrodynamic behavior of the reactor was represented as a plug-flow reactor, and the LiCl was the preferable tracer in this study.

Keywords: Bio-electrochemical reactor; hydrodynamics; RTD; plug-flow

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# **INTRODUCTION**

Researchers among different biological systemshave focused on biofilm systems, which are favourable for water and wastewater treatment. Fixed-bed reactors have been widely used due to simple operation, smaller reactor volumes, less sludge, greater stability, and capacity to handle shock loading [1]. Furthermore, high cell retention time in this type of reactor makes high rate BNR possible for processes with low grow rate bacteria [2]. On the other hand, they need longer start up time and sophisticated process control; furthermore, they are subject to possible clogging as well[3]. To well understand applying the fixed-bed reactor in practice to treat wastewater, the hydrodynamic study has been considered at numerous investigations during the last decades [2, 4, 5]. The biocarrier as an important part of a fixed bed reactor plays a key role at the performance of this system. The specific surface area as an important factor of the supporting medium, in the high value increase the efficacy of system [3]. The fixed bed reactors as more complicated systems than conventional systems due to the diffusive character of the flow passing through them and the conversion and elimination processes of contaminant, have been considered to point out the hydrodynamic performance of this type of reactors. Therefore, to determine the design parameters such as average residence time, a good description of hydrodynamic behavior is necessary[3]. Clogging as a main problem of fixed bed reactor after long time operation well occur because of the growth of biomass on biocarrier, which has an effect on reactor performance. The dead zone as a result of clogging situation influence the retention time in reactor and the contact time between bacteria and substrate[6, 7]. In the field of water and wastewater treatment, water injection with high velocity and aeration has been considered as a solution to reduce clogging. The residence time distribution (RTD) of the aqueous solution is subject to study of hydrodynamic behaviors of a reactor containing fixed biofilm via using a tracer material [8]. Furthermore, researchers applied a model namely

"biodiffusion model" based on dispersed plug flow and diffusion of the tracer within the biofilm[2, 7, 9]. Retention time as an important parameter must be in concern during the performance of a reactor, which is determined by the mixing condition in bioreactor. The mixing has a key role in attaining efficient change of reactants in an aqueous solution treatment, which is described based on the RTD conception[10]. As mentioned in previous studies, the performance of reactor is affected by the biofilm adhesion and detachment rates and the liquid-biofilm mass transfer resistance, which they are depend on residence time [3]. With RTDs, analysis can demonstrate the flow pattern and point out the presence of dead zones and short circuits in the reactor. Furthermore, the achieved results of analysis of RTDs make possible modeling of fixed-bed reactors at different arrangements[11, 12]. In this study, the hydraulic behavior of a TCUBER was investigated empirically by interpreting the residence time distribution curves. In addition, the applied tracers were compared at the same carrier with each other. The achievedresults of hydrodynamic characteristics were used to modify or to confirm the designed BER.

# **MATERIALS AND METHODS**

## **Experimental apparatus**

The Laboratory-Scale reactor used in this researchis shown in Fig.1. It consists of TCUBR, built from plexiglass tubes, which was composed of two compartments with a height of 42cm and inner diameter of 10 cm. The bottom of the reactor is provided with a mesh plate stainless steel (type 316) as electrodes at the bottom of each column and screwed to the plates, 12 stainless steel rods (5 mm in diameter and 40 mm in height) to enhance the supply of electricity in the palm shell-GAC as anode and cathode zones. In order to obtain hydrodynamic data, the system was operated in two stages: non-BER and BER stage.







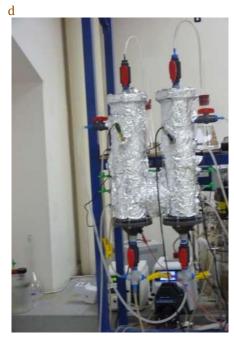


Fig. 1.Schematic of palm shell granular activated carbon twin chamber up-flow reactor (PSGAC-TCUR), when the carrier is sand(a), and GAC(b), stainless steel as electrode and flow disstributre(c), and when the reactor run as BER and carrier is GAC

The reactor employed as upflowfor three months, at four runs according to the amount of HRT (12, 24, and 48 h for non-BER and 12h when the reactor runs as BER), when other controlling parameters were constant (e.g. temperature=  $29\pm1^{\circ}$ C, PH =7.5±2 and current intensity only for BER=50mA). According to the result of the abiotic test, pH controlling is a necessary part to operate the system. Therefore, the pumparrond system has been used as a practicable method during the hydrodynamic study for both compartments. The effluent flow rate varied between 1.25 and5mL/min, and the re-circulating pump had a flow rate of around 20 mL/min in both columns. The operational conditions of system in both stages are summarized in Table 1.

Table 1. Operating Conditions for the Laboratory-Scale TCUBER

Experiments	Run 1	Run 2	Run 3	Run 4
HRT	6	12	24	6
Inlet flow (ml/min)	5	2.5	1.25	5
Recycling flow (ml/min)	20	20	20	20
Mixing speed(rpm) in lateral reactor	100	100	100	100
Temperature(ºC)	29±1	29±1	29±1	29±1
PH	7.5±0.2	7.5±0.2	7.5±0.2	7.5±0.2
Current (mA)	-	-	-	50

## **Inoculating and feeding**

The study of hydrodynamic behavior of reactor during actual operation was conducted for one run; acclimatized nitrifying and denitrifying biomass, from a local municipal wastewater treatmentplant (WWTP) was used to grow on the PSGAC. The initial total suspended solid (TSS) was 3000mgL<sup>-1</sup>. The composition of synthetic wastewater according to Table 2was injected to oxic part (anode compartment). The anoxic part (cathode compartments) was fed with effluent from the first compartment with the same influent.

Table 2. Synthetic wastewater compositions

NH <sub>4</sub> -N	200			
NaHCO <sub>3</sub>	C/N=8			
KH <sub>2</sub> PO <sub>4</sub>	N/P=5			
MgSO4 (g/l)	60			
рН	8			
Composition of trace element solution				
(1ml/L)				
EDTA	10			
ZnSO <sub>4</sub> .7H <sub>2</sub> O	2.2			
CoCL <sub>2</sub> .6H <sub>2</sub> O	3.2			
MnCl <sub>2</sub> .4H <sub>2</sub> O	10.2			
CuSO <sub>4</sub> .5H <sub>2</sub> O	0.22			
$(NH_4)_6Mo_7O_{24}\cdot 4H_2O$	2.2			
CaCl <sub>2</sub> .2H <sub>2</sub> O	1.1			
FeSO <sub>4</sub> .7H <sub>2</sub> O	10			
H <sub>3</sub> BO <sub>3</sub>	0.3			
NiSO <sub>4</sub> .6H <sub>2</sub> O	1			

# **Analytical methods**

The samples were analyzed immediately or were stored at cold temperature (4  $^{\circ}$ C) prior to the analysis. The samples for the determination of dissolved components, e.g., ammonium, nitrate, and nitrite concentration, were analyzed using an Advanced Compact Ion Chromatograph IC 861 (Metrohm® Ltd., Herisau, Switzerland) and guard column. The concentration of tracers Rodhamin-B dye belongs to the xanthene family and lithium ion in the effluent was measured using Spectroquant® pharo100 Merck and ICP-OES, OPTIMA300. Samples were filtered by filter 0.2 to avoid solids suspension. Temperature, pH, and DO were monitored continuously online in both stages of hydrodynamic studies. In addition, other experimental tests were applied using standard methods [13].

# Carrier support

In this experimental work, two types of carriers were used for packing reactor. In the first stage, river sand (2-4 mm) was used only as packing but in the second stage of the study, both compartments (anode and cathode) were packed by 600 g PSGAC from Bravo Green Sdn. Bhd., Sarawak, Malaysia with a size of 2 mm– 4 mm with a height of 22 cm as biocarrier and third electrode. The surface area of the PSGAC was determined as  $780 \text{ m}^2/\text{g}$  and the total pore volume was  $3.86 \text{ Cm}^3/\text{g}$ .

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## Tracer study

The stimulus-response method was applied for investigation of RTDs with a pulse or  $\delta$  –Direct injection (by a syringe over as short a time as possible (7 s)) of tracer from inlet of reactor at t = 0 and the concentration of the tracer (Ct) was measured by taking samples from the outlet of both compartments at regularly spaced intervals. The duration of each run was measured when the concentration of tracer was less than 5%, or more than 5 $\tau$ , where  $\tau$  is defined as theoretical average retention time according to equation(1).

Where V is the reactor volume and Q is the volumetric flow rate (equation (1)).

$$\tau = \frac{V_{\rm L}}{Q} \tag{1}$$

The time lapse between the two samples was as follows: every 30 min during the first 12 h, every 1 h during the following 12 h, every 2 h during the following 24 h, every 4 h during the following 24 h, and at the end the interval was 8h. The RTD profiles, which explain quantitatively how long a portion of the fluid has spent in the system, measured by determining tracer concentration in effluent, the outlet time distribution for a pulse input, characterized the RTDs of the fluid, E(t), defined according to equation (2)[2]:

$$E(t) = \frac{C(t)}{\frac{Q}{V_L} \int_0^\infty C(t) dt} = \frac{C_i}{\frac{Q}{V_L} \sum C_i \times \Delta t_i}$$
(2)

Researchers usually have applied momentums to compare RTD; the momentum technique allows the attaining of the n momentum of a RTD function (equation 3).

$$m_n = \int_0^\infty t^n E(t) dt \tag{3}$$

The first moment of the RTD function as experimental mean residence time (tm) equation (4) is known as average time that the molecules of the effluent remained inside the reactor. This can also be surmised as equation (5).

$$t_{m} = \frac{\int_{0}^{\infty} tC(t)dt}{\int_{0}^{\infty} C(t)dt}$$

$$t_{m} = \int_{0}^{\infty} tE(t)dt$$
(5)

The second moment of the function E(t) as variance,  $6^2t$  equation (6) was carried out to estimate the axial dispersion coefficient that indicates the spread of the RTD curve.

$$\sigma^2 = \int_0^\infty (t - t_m)^2 E(t) dt \qquad (6)$$

The skewness,  $s^3$ , of the curve of RTD as the third moment explains the extent that the distribution skews in one direction or the other, equation (7).

$$s^{3} = \int_{0}^{\infty} (t - t_{m})^{3} E(t) dt$$
 (7)

## RESULTS AND DISCUSSION

## Evaluation and analysis of hydrodynamic conditions

Standard curve for Rhodamine B dye was carried out by preparing standard solution in range of 0.2 to 5mg/L with deionized water. Representative absorption spectra of Rhodamine B in water are shown in Fig.2, in which the optimum wavelength for measuring the adsorption of samples was 554nm. The achieved maximum adsorption wavelength was applied for preparing the standard curve according to Fig.3.

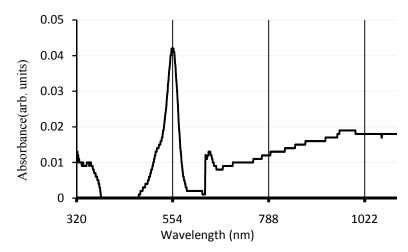


Fig. 2. Absorption spectra of Rhodamine B in water

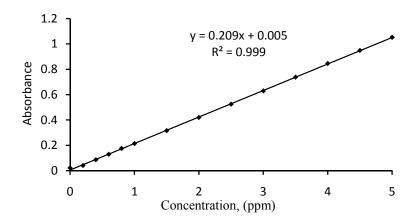


Fig. 3. Calibration Curve from 0.2 to 5 ppm of Rhodamin-B at 554 nm.

The results of tracer study to carry out hydraulic characteristics of the reactor based on RTD used normalized timeaccording to equation (8).  $\theta = \frac{t}{t_{\rm m}}$ 

$$\theta = \frac{\epsilon}{t_{\rm m}}$$
 (8)

On the other hand, RTD and its second and third moments are represented based on normalized time according to equations (9)-(11), where E(t) as function of a dimensionless time can be represented according to equation (9).

$$E_{\theta}(\theta) = t_{\rm m}E(t) \tag{9}$$

$$\sigma_{\theta}^2 = \int_0^\infty (\theta - 1)^2 E_{\theta}(\theta) \, d\theta = \frac{\sigma^2}{t_{\rm m}^2}$$

$$s_{\theta}^{3} = \int_{0}^{\infty} (\theta - 1)^{3} E_{\theta}(\theta) d\theta = \frac{s^{3}}{t_{m}^{3}}$$
 (11)

## Residence time distribution

The experiments were carried out at three different liquid flow rates (1.25ml/min, 2.5 ml/min, 5ml/min) twice. The almost complete recovery of tracers was considered during all runs. The demonstrated plots in Fig.4show the RTD curves obtained for the three runs with different flowrates as mentioned above. The plotted RTD curve for the first stage of hydrodynamic study when the reactor was applied as non-BER shows in all runs the flow in the system is close to the plug flow due to obtained result from  $t_p/\tau$  index, which is 1 or close to 1. However, the Dead Zone Index did not show significant results (the results were

close to zero) but the long tail on the tracer concentration curve on normalized time curve is due to dead zone. On the other hand, in the second stage, when the reactor worked as BER due to the production of oxygen and hydrogen gas in anode and cathode compartments respectively, the RTD curve is quite different from the results of the first stage and the mixing efficiency increased. In this stage because of some shortcomings such as adsorption of tracers, installation of permeable membrane in side reactor, and electrochemical reduction of tracers, the results of measured tracers were not suitable, and they are not shown here. However, according to Fig.4 in the first stage of experiments, when Rodamine-B was applied as tracer, the recovery was not the same as lithium due to porous packed media and adhesion of Rodamine-B. The peck concentration of lithium showed high ratio of recovery than Rodamine-B.

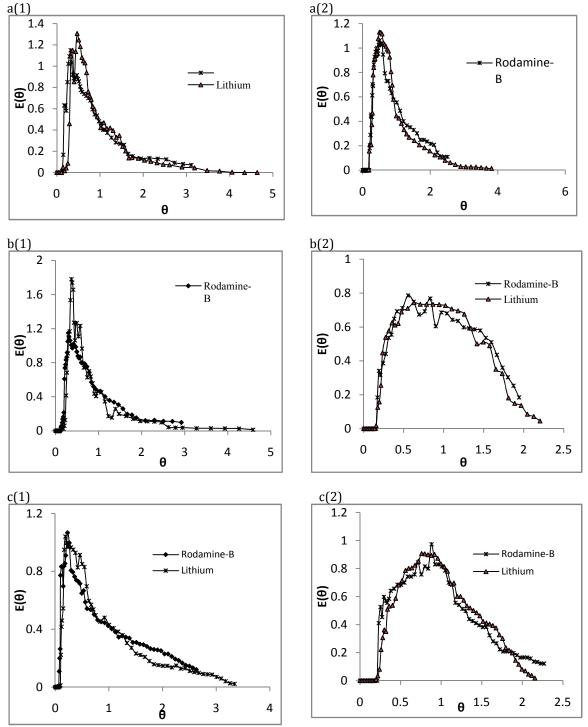


Fig.4. Experimental curves of normalized residence time distribution(RTD) from the anode and cathode compartment in a TCUBER. Condition: (a) HRT(12h); (1)anode (2) cathode, (b) HRT(24h); (1)anode (2) cathode

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## **CONCLUSION**

The study of hydrodynamic characteristics of PSGAC-TCUBER as BER and non-BER, demonstrated different behaviors, due to the effect of operational parameters(e.g. current intensity, HRT) and type of packing. Furthermore, applied tracers (R-B, R-S) showed different results; LiCl as the preferable tracer established reasonable results during the study, however, R-B due to adhesion on the surface of the reactor and fitness, as well as adsorption by packing material specially PS, gives unsuitable results in both stages of the study. The result demonstrated a plug flow behavior for the reactor specially when the flow rate increased to a high amount (48h), with small dispersion values, and dead zones in both side significantly decreased. In addition, achieved results confirmed that the HRT affected the hydrodynamic behavior of TCUBR.

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