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Electrochemical regeneration of granular activated carbon saturated with Rhodamine B in a fluidized electrochemical reactor

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ABSTRACT

This study investigated the electrochemical regeneration in fluidized reactor, used for granular activated carbon (GAC) saturated with Rhodamine B (RB). The regeneration efficiency is mainly affected by the generation of active spices including hydroxyl radicals. In this present modified study, the improvement of mass transfer using fluidized reactor lead to increase regeneration efficiency. This process was modelled and optimized using response surface methodology (RSM) based on central composite design (CCD). Such effective parameters on regeneration efficiency (RE) as current density, regeneration time and electrolyte flow rate were optimized and modelled using reduced quadratic model. The predictive ability of proposed model was verified by computing correlation coefficient ($R^2 = 0.9956$). At optimum condition, effective parameters of current density = 3.79 (mA cm^{-2}), regeneration time = 100 (min) and electrolyte flow rate = 274.5 (mL min^{-1}), maximum regeneration efficiency (83.7%) was achieved. The results of which were obtained after four time sequential adsorption-desorption cycles were acceptable. In conclusion, the modified electrochemical regeneration method is suggested to be used more widely as cost-effective technique.

Keywords: Electrochemical regeneration; Granular activated carbon; Fluidized reactor; Central composite design (CCD); Rhodamine B.

1. Introduction

Adsorption due to its high efficiency and low cost, is preferred as compared to other methods for removal pollutants, widely used in many industries. Among the different types of adsorbents, activated carbon due to its high surface area and pore distribution has been employed on a large scale for wastewater treatment [1]. But activated carbon is relatively expensive and easily saturated, thus the use of GAC without regeneration would not be economic and causes environmental pollution, therefor activated carbon regeneration is essential to economic and environmental reasons [2].

It is noteworthy that there are various techniques employed for regenerating of GAC including thermal [3], chemical [4], photo-assisted [5], electrochemical [6], ultrasound [7] and dielectric barrier discharge plasma (DBDP) [8]. Among the techniques mentioned, thermal regeneration is believed to be the most common method but has some drawbacks such as high energy requirement and GAC loss [9]. Chemical regeneration of GAC is widely used with high efficiency, however this method is high cost and its regeneration efficiency depends on types of pollutants [10]. Photo-assisted oxidation has the merit of fast degradation of organic pollutants to very low concentration, but there are no evidences to prove that intermediates from process are not toxic to human especially from chlorinated pollutants. DBDP regeneration technique is a recent tool that has been proven to be very efficient. However, the problem of harmful by-product such as tri- and tetrachlorophenols, chloride, glyoxalic acid, formic acid, maleic acid, etc. being formed in the process, is main drawback suffered by this technique. Recently ultrasonic regeneration attracted the attention of several researchers. The effect of ultrasonic irradiation is revealed through the cavitation phenomenon. Some advantages of this technique are no attrition of GAC, its low power consumption, simpler process equipment and higher desorption efficiency [11].

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Recently, electrochemical regeneration has been reported to be a promising approach. The uniqueness of this method including environmental adaptability, selectivity, no addition chemical and cost effectiveness [12]. Numerous studies have been performed to employ this technique [13,14]. Narbaitz and Cen have studied the electrochemical regeneration GAC saturated with phenol and achieved more than 95% regeneration efficiency [15]. Zhang investigated electrochemical regeneration of GAC saturated with phenol and reported more than 80% regeneration efficiency was obtained [16].

Zhou and Lei studied the electrochemical regeneration of activated carbon loaded with P-nitrophenol in a fluidized rector and obtained around 90% regeneration efficiency [17].

However, long time to obtain high efficiency is required to reach high regeneration efficiency, which does influence the economics of the process and then limits its application. The performance of electrochemical regeneration depends on two factors:

- Electrode material has important role in the electrochemical oxidation, modified electrodes could generate more activated spices such as hydroxyl radicals [18].
- Mass transfer could play key role in regeneration time and economics of the method. Degradation of pollutants more likely occurs on the surface of the electrodes rather than the bulk of the solution, so that degradation intermediates remain on the electrode which led to the electrode fouling [19], that is defined as the blocking of the electrode surface by intermediates produced during oxidation [20].This phenomenon reduced regeneration efficiency and increased the regeneration time. Therefore, modification of this process is essential. In the modified method, the time-space efficiency for organic pollutants degradation is enhanced by promoting mass transfer, so it could reduce the regeneration time [21].

By considering these problems and in order to promote the efficiency of regeneration, in the current study the improvement of mass transfer is considered in a novel fluidized reactor. As previously described, electrochemical oxidation has a main role in degradation of desorbed pollutants, this process was found accelerated by considering mass transfer in a novel fluidized reactor.

The synergic effect of electrochemical oxidation and promoted mass transfer has been investigated in this project to verify its advantages. In this reactor, regeneration of GAC saturated with Rhodamine B was investigated. Rhodamine B is one of the Synthetic dyestuffs used by several colorant manufacturing such as textile industries, considered as a class of anthropogenic pollutants causes serious problems to water environment [22]. Rhodamine B, due to the presence of N-ethyl groups at either side of xanthene ring can cause several damage to human beings and environment [23]. Taking the impact of effluent on environment into consideration, treatment of waste water containing such compound, is deemed essential [24].

2. Experimental

2.1. Chemical and materials

Purchased GAC from shimi co., Hamedan, was prepared from walnut shell. Then, GAC was washed several times by deionized water followed by drying for 5 h at 100ºC to a constant weight. Before regeneration, saturation of GAC must be carried out. For saturating, 0.5 g of GAC was soaked with 200 mL RB (170 mg L^{-1}), shaken for 24 h, at room temperature to ensure the adsorption/desorption equilibrium. UV-spectrophotometer (Jasco-V630) was used for the analysis of remaining RB concentration. This measurement was performed to evaluate adsorption capacity of GAC and to ensure the fact that saturation of GAS has completely occurred. Then saturated GAC was separated out and was dried for 5 h at 100ºC storing the next distinct electrochemical regeneration.

2.2. Adsorption isotherm for RB

Adsorption experiments were conducted to evaluate capacity of GAC and to ensure that GAC was totally saturated. Measuring of adsorption capacity was carried out by using many flasks with equal volume (200 mL) and different initial concentration of RB around (40–300 mg L^{-1}), A certain amount of GAC with same weight (0.3 g) was put into flasks. The flasks were shaken at room temperature for 24 h to reach equilibrium. The amount of RB adsorbed on GAC at equilibrium can be calculated by using fallowing equation:

$$q_e = \frac{V}{M}\left(C_0 - C_e\right) \tag{1}$$

where C_0 and C_e are initial and equilibrium concentration of RB (mg L^{-1}) respectively, those are determined by UV-spectrometer, V is the volume of solution and M is weight of GAC. q is the amount of RB adsorbed onto GAC, measured as RB per gram of GAC. Adsorption isotherm of RB is shown in Fig. 1.

2.3. Electrochemical regeneration

The schematic of electrochemical reactor used for regenerating GAC is presented in Fig. 2. This reactor consisted of two electrodes, titanium mesh anode (1 cm × 1 cm) and

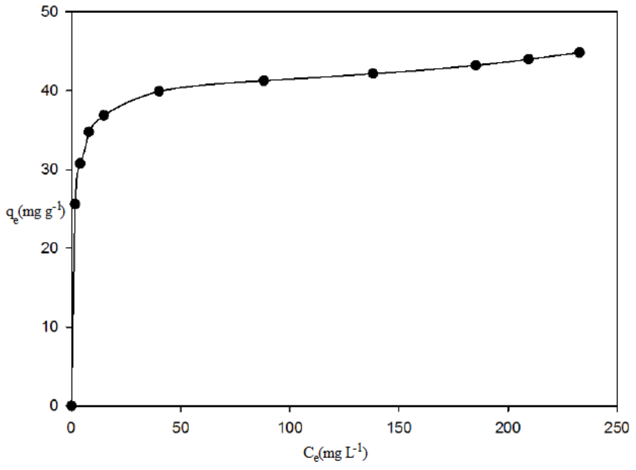

Fig. 1. Adsorption isotherms of fresh GAC.

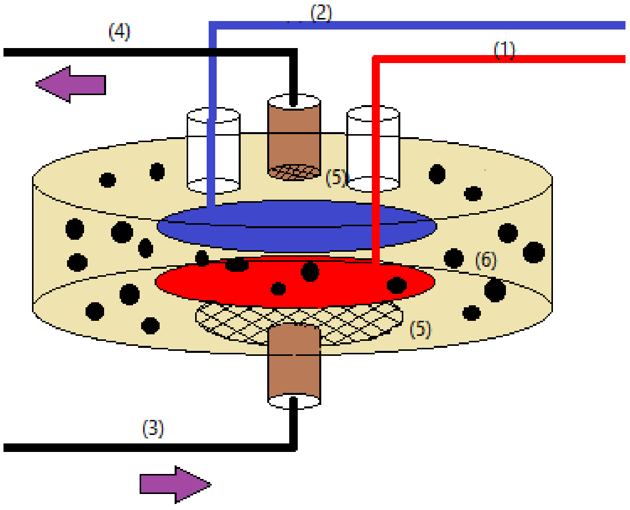

Fig. 2. The schematic diagram of the fluidized electrochemical reactor. (1) anode, (2) cathode, (3) inlet, (4) outlet, (5) filter, (6) saturated GAC.

stainless steel cathode (3 cm × 2 cm), Na_2SO_4 was used as electrolyte (500 mL, 0.1 M). A peristaltic pump (ISMATEC-MCP) used for circulating and measuring the electrolyte flow rate. DC source (BK-PRECISION) applied desired constant current density to the electrodes.

The effective range of regeneration time would be 45–150 min, so this parameter has been investigated in this range. After electrochemical process, regenerated GAC was filtered and put into flasks for 24 h to assess re-adsorption capacity. The comparison of adsorption capacity of fresh GAC and re-adsorption capacity of regenerated GAC demonstrates the effectiveness of regeneration process.

2.4. Analysis

Concentration of RB in different solution was identified by using calibration curve, which was plotted based on Lambert-beer law at $\lambda_{max} = 554$ nm. The percentage of adsorption capacity of GAC (fresh or regenerated) was also calculated using this formula:

$$\%\mathrm{ads} = \frac{C_0 - C_e}{C_0} \times 100 \tag{2}$$

In this equation C_0 and C_e are initial and equilibrium concentration of RB (mg L^{-1}) respectively. Dividing adsorption capacity of fresh GAC on re-adsorption capacity of regenerated GAC multiplied by 100 indicates the regeneration efficiency percentage.

$$\%\mathrm{RE} = \frac{\%\mathrm{ads}(\mathrm{fresh})}{\%\mathrm{ads}(\mathrm{regenerate})} \times 100 \tag{3}$$

Structural changes of GAC during electrochemical regeneration were investigated by BET test. The surface area and pore distribution GAC (fresh or regenerated) was determined using BET equation.

2.5. Experimental design

Response surface modeling is an empirical statistical technique that determines regression model and operation condition by using quantitate data obtained from designed experiments [25].

The individual and cumulative effect of considered variables and subsequent effects on response were described by graphical representation. The main aim of RSM is to determine optimal condition for process. In order to reduce process variability and experimental time, employment of statistical experimental designing is essential [26].

The most popular class of second-order design called central composite design was used in experimental design. In CCD, the examination of the effective variables and the interactions between them was conducted by minimum number of experiments. In our study, RE% was considered as a response and function of main variables, including electrolyte flow rate, current density and regeneration time. Electrochemical regeneration of GAC saturated with RB was optimized by RSM based on CCD.

3. Result and discussion

The experimental design was carried out by RSM based on CCD, which is the most popular second-order design. The empirical relationship between RE%, as response surface, and independent variables including current density (X_1), regeneration time (X_2) and electrolyte flow rate (X_3) was presented by using CCD. In this study, three-factor and five level CCD was applied for optimizing electrochemical regeneration of GAC saturated with RB.

Preliminary experiment was carried out to determine the ranges of independent variables which were assessed at 5 levels between $-\alpha$ and $+\alpha$. The levels of effective variables considered in this study, were presented in Table 1.

According to ($2^n + 2n + n_c$), where n is the number of independent factors, the total number of experiments is given as sum of the 2^n factorial runs, 2n axial runs and n_c centre runs. For three factor process, a total of 20 experiments consisted of 8 factorial points, 6 axial points and 6 replicates at centre point were performed. In each experiment, conditions were adjusted based on the experimental design matrix which is displayed in Table 2. RE%, which was taken as response of experimental data, was analysed to evaluate performance of the process.

For optimization the process, the main variables in the electrochemical regeneration were considered as a function of response by quadratic model. A quadratic model was given in Eq. (4):

$$Y = \beta_0 + \sum\nolimits_{j=1}^{k} \beta_j X_j + \sum\nolimits_{j=1}^{k} \beta_{jj} X_j^2 + \sum_i \sum\nolimits_{<j=2}^{k} \beta_{ij} X_i X_j + e_i \tag{4}$$

where Y represents predicted response (RE %), X_i and X_j are effective factors, β_0 is the constant term, β_j is the linear coefficient, β_{jj} is quadratic coefficient and β_{ij} represents interaction coefficient. K is number of variables and e_i is the error.

Design Expert 7.1.3 software was employed to analyse and process the experimental results. Analysis of variance (ANOVA) provide a collection of diagnostic tests which evaluate adequacy of the proposed model. The predictive

Table 1
The experimental ranges and levels of independent variables in terms of real and codded factors

	−α	−1	0	+1	+α
Current density (mA cm^{-2}), X_1	1	1.81	3	4.19	5
Regeneration time (min), X_2	45	66	96	135	150
Electrolyte flow rate (mL min^{-1}), X_3	100	181.08	300	418.92	500

Table 2
The 3-factrors central composite design matrix for regeneration efficiency (RE %) and observed response

Experimental run	Current density (mA cm^{-2})	Regeneration time (min)	Electrolyte flow rate (mL min^{-1})	Regeneration of GAC (%)
1	4.19	128	181.08	73.5
2	3.00	97	300.00	81.5
3	4.19	66	418.92	71.9
4	3.00	97	300.00	82.9
5	1.81	128	181.08	67.7
6	4.19	128	418.92	72.7
7	1.81	128	418.92	68.2
8	3.00	97	100.00	65.6
9	1.81	66	418.92	55.2
10	3.00	97	500.00	58.8
11	1.00	97	300.00	61.2
12	5.00	97	300.00	80.4
13	3.00	45	300.00	72.4
14	3.00	97	300.00	83.4
15	1.00	66	181.08	62.3
16	3.00	97	300.00	82.6
17	3.00	97	300.00	81.3
18	3.00	97	300.00	80.8
19	3.00	150	300.00	70.4
20	4.19	66	181.08	79.8

ability of proposed model was verified by computing Correlation coefficient (R^2) [27].

Adequate polynomial model displaying the empirical relation between response and variables, are given in Eq. (5):

$$\%RE = 82.08 + 5.62X_1 + 1.81X_2 - 1.91X_3 - 2.99X_1X_2 + 1.84X_2X_3 - 3.97X_1^2 - 2.16X_2^2 - 7.08X_3^2 \tag{5}$$

The terms with positive sign have synergic effect whereas negative sign indicates antagonistic effect. The effects of process variables on electrochemical regeneration were evaluated by using mentioned model.

ANOVA results (Table 3) of regression model confirm the significance and accuracy of proposed model. High value of coefficient of determination ($R^2 = 0.9956$) and (adjusted $R^2 = 0.9924$) confirm the agreement between obtained and predicted values. Desirable value of R^2 implies the relation between factors and response was explained satisfactorily by this regression model [28].

The proposed model also was evaluated by residual, which is defined as the difference between experimental and predicted response values. A good mathematical model fitted to experimental must present low residual values. The lack of fit was non-significant and desirable. The non-significant lack of fit implies the quadratic model was satisfactorily fitted to the experimental responses.

3.1. Interactive effect of electrolyte flow rate and current density

The combined effect of electrolyte flow rate and current density on regeneration efficiency at constant regeneration time was investigated and is illustrated in Fig. 3. Graphs show that the maximum regeneration efficiency occurs at the current density of 3.79 mA cm^{-2} and the electrolyte flow rate of 274.5 mL min^{-1}, which is in accordance with the model. It is evident that RE% increases with the increase of electrolyte flow rate until 300 mL min^{-1} but regeneration efficiency started to decrease with further increase in flow rate.

The electrolyte flow rate affects mass transfer significantly, which may in turn influences the electrochemical oxidation and desorption. Thus, it would influence the electrochemical efficiency.

At lower flow rate, GAC was fixed. When flow rate was increased to 240 mL min^{-1}, GAC just started to fluidize. After fluidization of GAC, the enhancement of flow rate increased the regeneration efficiency. However, RE% decreases with increasing of flow rate. The observed trend may be understood that, at highest flow rate, the residence time in the system became too short that hydroxyl radicals couldn’t degrade pollutants effectively. So in this study, result implies that the optimal flow rate for electrochemical regeneration was 274.5 mL min^{-1}.

In another experiment, exhausted GAC was investigated under different flow rates at the same time without current charged. In this study, only desorption of saturated GAC has occurred and found that the RE% under different flow rate were all among 9–12% . The result of this experiment indicated that electrochemical oxidation plays principle role in the GAC regeneration and also revealed the reason for increasing of RE% at high flow rate.

The adsorbed pollutant on the GAC could not be directly degraded by active spices such as hydroxyl radicals, those are at or very near the anode [29], also the life span of hydroxyl radicals are so short. Therefore, they wouldn’t diffuse through the bulk solution onto the GAC. Electrochemical regeneration of saturated GAC followed electrodesorption mechanism [15]. In this mechanism, two factors including electrochemical oxidation and desorption play key role in regeneration process [30]. Based on this mechanism, The RB adsorbed would firstly be desorbed from GAC and then be shifted to the surface of the anode to be further oxidized by electrochemical oxidation. An improved mass transfer of the desorbed RB to or close to the anode lead to higher the RE% of the exhausted GAC. Also it might speed up the GAC

Table 3
ANOVA for fit of regeneration efficiency (RE %)

Source of variation	Sum of square	Degree of freedom	Mean square	F value	Prob > F	
Model	1521.66	8	190.20	313.03	<0.0001	Significant
Lake of fit	1.42	6	0.24	0.22	0.9518	Non-significant
Pure error	5.27	5	1.05			
Residual	6.68	11	0.61			
Core total	1528.29	19				

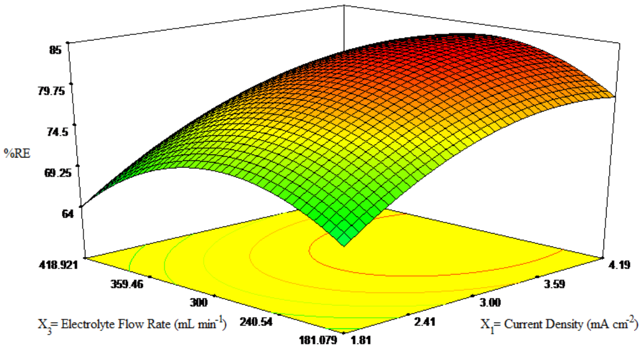

Fig. 3. Surface response for the regeneration efficiency (RE %) as a function of current density and electrolyte flow rate.

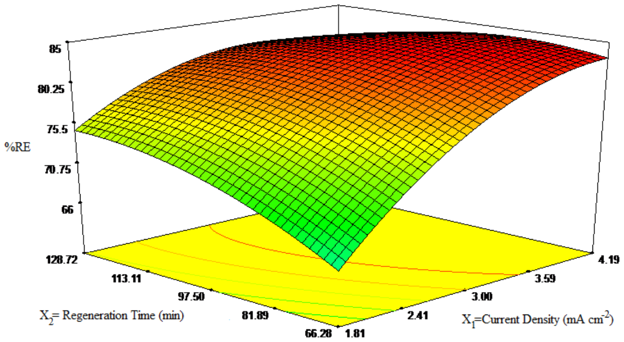

Fig. 4. Surface response for the regeneration efficiency (RE %) as a function of regeneration time and current density.

regeneration. Similar explanation have been presented by others [15,16,21].

3.2. Interactive effect of current density and regeneration time

The effect of current density and regeneration time was analysed as shown in Fig. 4 from CCD and it has been estimated that the point of maximum RE% of GAC was achieved by using the combination of two process variables, i.e. current density of 3.79 mA cm^{-2} and regeneration time of 100 min. It is evident that electrochemical oxidation process is significantly influenced by current density. As previously described, it was found that in the absence of current density RE% were all among 9–12%. The regeneration of GAC should be attributed to the current density and the contribution of desorption could be omitted. Degradation mechanism of RB has been proved mainly due to electrochemical generation of hydroxyl radicals as follows [31]:

$$H_2O \rightarrow OH^{\bullet} + H^{+} + e^{-} \qquad (6)$$

RE% increases with increase of current density until 3.79 mA cm^{-2} but decreases with further enhancement of applied current density, this might be attributed to the destruction of pores due to the oxidation of GAC at higher current density, which in turn, lead to decrease pores volume, adsorption ability of GAC and regeneration efficiency.

Regeneration time is another factor affecting economics of the process. RE% increases with regeneration time but no evident enhancement was observed after being treated 100 min. Thus,100 min would be optimal time for GAC regeneration.

3.3. Regeneration cycles

The repetitive regeneration of GAC under optimum condition was performed in order to investigate the effect of regeneration cycles. It should be taken into consideration that reusing times affect economical aspect of the process. Therefore, the assessment of regeneration times is essential. In this experiment, GAC was saturated and regenerated electrochemically under the obtained optimized condition, repeatedly. As can be seen from Fig. 5, RE% decreased with the increase regeneration times, which may be attributed to the decrease of micro pore volume, which, in turn, could decline the capacity of adsorption of GAC and RE%. However, the decrease of RE% after four times regeneration was insignificance. In other words, the structure of pores was not changed during Sono-Fenton regeneration.

3.4. Process optimization

The main aim of this study is to achieve maximum RE% and minimum energy consumption. The optimum values of three main variables were determined and the confirmatory experimental run under optimum condition showed a regeneration efficiency of 83.7%, this close agreement confirm the model weight. Table 4 presents optimum values and the predicted and observed of regeneration efficiency.

The surface area of fresh, saturated and electrochemical regenerated GAC under optimum condition was calculated using BET equation. The results indicated the surface area decreases from 840 to 483 m^2 g^{-1} during saturation process. The t-method determines specific area of meso pores and micro pores. The results showed that surface

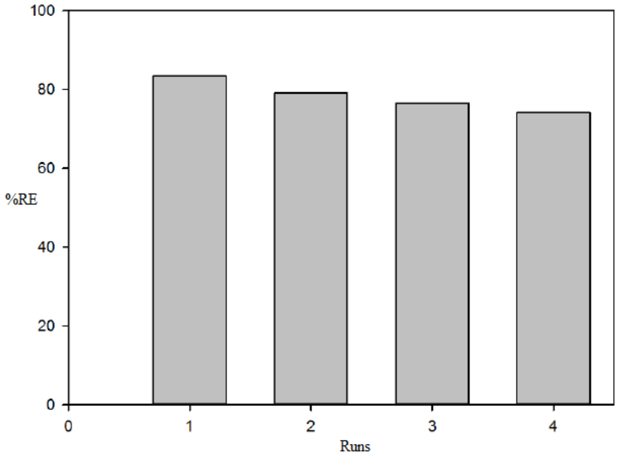

Fig. 5. Regeneration efficiency percentage during four times regeneration.

Table 4
Optimum values of regeneration efficiency of GAC saturated with RB

Current density (mA cm^{-2})	3.79
Regeneration time (min)	100
Electrolyte flow rate (mL min^{-1})	274.5
Predicted RE%	84.08
Observed RE%	83.7

area of micro pores significantly decreases in saturated GAC. In other word, the adsorption of RB has occurred in micro pores. During the regeneration of GAC under optimum condition, BET surface of GAC and specific area of micro pores increase which demonstrated that RB was transferred and degraded. The surface area of regenerated GAC was 793 $m^2 g^{-1}$. The calculated RE% through the consideration of surface area of fresh, saturated and regenerated GAC was obtained around 86.83%.

4. Conclusion

Electrochemical regeneration of GAC saturated with RB in a fluidized electro-reactor has been investigated. The combination of electrochemical oxidation and promoted mass transfer has been proved to be a suitable method. The effect of three main variables including current density, regeneration time and electrolyte flow rate on RE% were evaluated. It was found that electrochemical oxidation was an principle reason for GAC regeneration, but improvement of mass transfer using fluidized reactor could promote this process and increase RE%. From the optimization, maximum regeneration efficiency (83.7%) at current density of 3.79 mA cm^{-2}, regeneration time of 100 min and electrolyte flow rate of 274.5 mL min^{-1} were achieved. Energy cost would also be evaluated, the calculated energy consumption was around 0.363 kWh kg^{-1}, which demonstrates this method is cost-effective. Moreover, insignificant decrease of RE% was observed after four times regeneration. ANOVA results showed high value of determination coefficient ($R^2 = 0.9956$) and (adjusted $R^2 = 0.9924$). Thus, ensuring a satisfactory adjustment of the second order regression model with the experimental date. Hence, the combination of electrochemical oxidation and promoted mass transfer in a novel fluidized electro-reactor represents a suitable technique to improve the efficiency of regeneration.

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