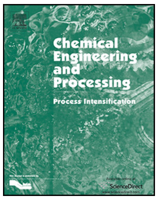
Chemical Engineering and Processing 105 (2016) 92–102

Contents lists available at ScienceDirect

Chemical Engineering and Processing:
Process Intensification

journal homepage: www.elsevier.com/locate/cep



Integrated ozone—electrocoagulation process for the removal of pollutant from industrial effluent: Optimization through response surface methodology

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A R T I C L E I N F O

Article history:
Received 20 December 2015
Received in revised form 14 March 2016
Accepted 17 March 2016
Available online 18 March 2016

Keywords:
Ozone assisted electrocoagulation
Distillery effluent
Central composite design
Color and COD removal
Power consumption

A B S T R A C T

In this study, the efficiency of ozonation, electrocoagulation and ozone assisted electrocoagulation processes for removal of pollutants from effluent of distillery industry was compared. The experimental results showed that ozone assisted electrocoagulation process yielded higher pollutant removal than ozone and electrocoagulation processes alone. Response surface methodology based on central composite design was used to optimize various operating parameters of the ozone assisted electrocoagulation process for the treatment of industrial effluent. The effects of five independent parameters such as current density(X_1), COD concentration(X_2), effluent pH(X_3), inter-electrode distance (X_4) and electrolysis time(X_5) on the% COD removal and power consumption were investigated. A quadratic model was used to predict the COD removal and power consumption in different conditions. The significance of independent variables and their interaction were evaluated by ANOVA. In order to achieve the maximum COD removal and minimum power consumption, the optimum conditions were obtained by mathematical and statistical methods. The results showed that maximum COD removal efficiency could be achieved at optimum conditions of $X_1 - 3A/dm^2$, $X_2 - 3000$ ppm, $X_3 - 7$, $X_4 - 1.8$ cm and $X_5 - 5$ h. In conclusion, hybrid electrocoagulation process could be applied successfully for removing pollutants from effluent.

1. Introduction

Across the world, there continues to be a large volume of wastewater that is generated daily from different industries and facilities such as leather industry [1], pulp and paper [2], petrochemical [3], text tile [4,5], distillery [6], pharmaceutical [7], hospital and slaughterhouse [8] and municipal wastewater treatment facility [9] etc. Industrial wastewater usually contains pathogens, persistent contaminants [10], pesticides [11–13] and heavy metals [14–16] such as arsenic, chromium, copper, boron and nickel [17–19], fluoride[20] etc. If the industrial effluent and wastewater is not properly treated before being discharged directly into rivers, water bodies, environment, human health and different organisms can be negatively affected. Water pollution caused by industrial wastewater leads to oxygen depletion in water bodies. Such pollution also causes beach closures, disrupted ecobalance, restrictions on recreational water use, reduction in shellfish harvest and contamination of drinking water. Currently, the molasses based distillery industry is considered one of the most polluting industries in the world. The effluent generated from the distillery industry has a high concentration of decomposable organic matters and dissolved salts. It also has a persistent dark brown color. Different treatment techniques are available for the removal of pollutants from the distillery industry such as physical [21,22], chemical [23] and biological process [24]. Biological treatment process is less expensive than the other methods. However, biological treatment is ineffective for color removal because the distillery industry effluent has persistent dark brown color. During the physical treatment process, contaminants in effluent are transformed from one phase to another and further treatment is needed. Therefore, there is a need to develop novel methods to overcome the above difficulties. Over the last few decades, electrochemical and advanced oxidation processes (AOPs) have been widely used for removal of a broad range of organic and inorganic pollutants from the various industrial effluents [25–28]. Simple electrochemical and/or advanced oxidation processes are not able to substantially mineralize organics and inorganics present in industrial effluent [29]. Coupling AOPs with other treatment process

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http://dx.doi.org/10.1016/j.cep.2016.03.013
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such as electrochemical technologies is effective to achieve high organic and inorganic removal efficiency with requirement of lower energy consumption at minimal cost. Several researchers have focused on the application of hybrid electrochemical processes such as the photo-electrochemical, ozone-electrochemical, sono-electrochemical process to remove pollutants from various industries [30–34]. Bernal-Martínez et al. reported that effectiveness of the electrochemical, ozone and integrated electrochemical–ozone processes for the treatment of industrial park wastewater and they found that the electrochemical–ozone process significantly improved the removal of COD, BOD_5, color, turbidity and total coliforms [30]. Hernández-Ortega et al. treated chemical industry wastewater by electrocoagulation and ozonation pre-treatments to further enhance efficiency of the existing biological treatment. They found that biological treatment alone was not effective for treating raw effluents, but high-quality treated effluent was achievable when biological treatment was combined with the electrocoagulation–ozonation process [35]. Bernal-Martínez et al. studied the performance of the electrochemical, ozone and integrated electrochemical–ozone processes for industrial wastewater treatment and they are reported that, the combining the electrochemical–ozone process with energy pulses was a suitable process for wastewater treatment because the amount of sludge and electrodes passivation was lower than that of the electrochemical process alone [36]. The hybrid electrochemical treatment process is an eco-friendly and cost-effective method. The pollutant removal mechanisms during the hybrid electrocoagulation process include coagulation, adsorption, precipitation and flotation. The hybrid electrochemical process has several advantages over single electrochemical process such as shorter reaction time, less sludge production, high efficiency, and capability to combine with other treatment processes, low cost, easy operation and more effective in purifying water [37]. Based on the literature review, there are no scientific studies on the ozone assisted electrocoagulation process for the pollutants removal from real industrial effluent using response surface methodology (RSM). The main objectives of this research work are to develop a novel technology based on the combination of electrochemical and AOPs for treating the distillery industrial effluent and to obtain the process performance through studying various operating parameters using central composite design (CCD).

1.1. Response surface methodology (RSM)

The parameters chosen for the hybrid electrochemical process in this study were optimized statistically by adopting response surface methodology. RSM is a regression analysis used to predict the value of a dependent variable based on the controlled values of independent variables. It can generate a lot of combinations of experimental parameters within a short period of time to make laboratory tests more efficient. Using the estimated parameter, one can determine the variable that contributes the most to the estimated value, thereby allowing the researcher to focus on the variables that are the most important to product acceptance [38]. RSM is employed for modeling and optimizing different chemical [39,40], physical [40–42] and biological processes [43] for a variety of pollutants. The two mostly used designs in RSM are Central Composite Design (CCD) and Box Behnken Design (BBD). BBD is a collection of three-level designs that have various geometric constructions. CCD consists of (a) factorial points, which are a 2^k or resolution V 2^{k-f} design allows estimation of the linear and linear – by – linear term in general quadratic model; (b) star or axial points, which have each factor in turn set to its high and low levels and the other factors at their central and (c) center points, which have all factors set to their central level. CCD is ideal for sequential experiments and it allows a reasonable amount of information for testing the lack-of-fit using a reasonable number of design points. This method has been widely used for optimization studies in recent years.

In many technical fields, it is a common problem that the variable y (output variable) exists with a set of predictor variables x_1, x_2, . . . , x_k (input variables). It can be written as an empirical model $y = f(x_1, x_2, \ldots, x_k) + \varepsilon$, where, ε represents the error in the model, and *f* represents the unknown response surface. Generally, a first-order or second-order polynomial is deduced to describe *f*. This empirical model is called response surface model and the approximation of the response function $y = f(x_1, x_2, \ldots, x_k) + \varepsilon$ is called response surface methodology.

Either first-order or second-order polynomial models are employed in order to develop proper approximation for *f*.

In general, the first-order model is given below.

$$y = \beta_0 + \sum_{j=1}^{k} \beta_j X_j + \varepsilon \tag{1}$$

and the second-order model is s follows:

$$Y = \beta_0 + \sum_{j=1}^{k} \beta_j X_j + \sum_{j=1}^{k} \beta_{jj} X_j^2 + \sum_{j=1}^{k-1} \sum_{i=2}^{k} \beta_{ji} X_j X_i + \varepsilon_i \tag{2}$$

where, X_i, X_j are coded independent variables, β_j, β_{jj}, β_{ji}($i = 1, 2, \ldots, k$; $j = 1, 2, \ldots, k$) are the regression coefficients. The first-order model describes a flat surface. The second-order model describes a curving surface, including all terms in the first-order model, plus all quadratic terms like $\beta_{jj}x^2_j$, and all interaction terms like $\beta_{ji}x_jx_i$. The second-order model is also called quadratic model. This model is generally adequate for RSM in most cases. The independent variables are often called predictor variables or regressors, and therefore the first-order or second-order models are also called regression models. Furthermore, fitting an appropriate response surface model requires the use of statistical fundamentals, regression modeling techniques and optimization methods.

In the present investigation, the operating parameters of the ozone assisted electrocoagulation process such as current density (X_1), COD concentration (X_2), effluent pH (X_3), inter-electrode distance (X_4) and electrolysis time (X_5) were optimized by using

Table 1
Coded and actual values of the variables of the design of experiments for the overall hybrid electrocoagulation process.

Variable	Unit	Factors	Levels				
			−2	−1	0	+1	+2
Current density	A/dm^2	X_1	1	2	3	4	5
Effluent COD concentration	ppm	X_2	1000	2000	3000	4000	5000
Initial effluent pH	–	X_3	3	5	7	9	11
Inter-electrode distnace	cm	X_4	0.6	1.2	1.8	2.4	3.0
Electrolysis time	h	X_5	1	2	3	4	5