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Dairy wastewater treatment using an electrochemical method: Experimental and statistical study

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ABSTRACT

In this paper, a dairy wastewater treatment was performed by electro-Fenton process using iron electrode. The experiments were conducted to evaluate the effect of five important parameters including reaction time, current density, pH, H_2O_2/Fe^{2+} molar ratio and H_2O_2/Dairy wastewater (DW) (ml/l) on the performance of the process. The response surface methodology was applied to minimize the number of runs and investigate the optimum operating condition. Five independent variables were carefully considered and optimized in this research. The optimum conditions were reaction time of 90 min, current density of 56 mA/cm^2, pH of 7.52, H_2O_2/DW volumetric ratio of 0.898 ml/l and H_2O_2/Fe^{2+} molar ratio of 3.965 for 93.93% COD removal while the optimum conditions were reaction time of 86 min, current density of 55.1 mA/cm^2, pH of 7.48, H_2O_2/DW volumetric ratio of 0.907 ml/l and H_2O_2/Fe^{2+} molar ratio of 3.987 for 97.32% color removal.

1. Introduction

Dairy industry is one of the biggest water consumers between the food industries [1,2]. The wastewaters obtained from the dairy industries are characterized by large amounts of organic materials, suspended solids, oils, salts and fats [3]. The effluent depends on the amounts of processed milk, type of products and equipments, production method, system management and washing mechanism [4]. These wastewaters are characterized by high levels of Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD) and nutrient matters (nitrogen and phosphorous [5,6]). Dairy wastewater treatment is an important task due to environmental issues and water recovery [5].

There are several techniques for dairy wastewaters treatment. They are generally treated using physico-chemical and biological methods [6]. Chemical treatment can completely remove the contaminant agents from the dairy wastewaters [6,7]. Furthermore, there are the other treatment methods such as nanofiltration (NF) [8], reverse osmosis (RO) [8–10], coagulation and flocculation [5], electrochemical treatment [11,12], membrane bioreactors [13] and adsorption [14]. Biological methods such as activated sludge process, aerated lagoons, sequencing batch reactors (SBR), upflow anaerobic sludge blanket (UASB) and anaerobic filters need a wide area and long time treatment. They also generate a lot of sludge [6]. Advance oxidation processes (AOPs) have recently encouraged for wastewater treatment [15]. Fenton oxidation (FO) is one of the AOPs. Hydroxyl radical is one of the strongest oxidants in this process [16]. This process target is to generate hydroxyl radicals for the organic substances destruction. Hydroxyl radical is one of the free radicals most reactive which can be easily produced by a reaction between H_2O_2 and Fe^{2+} ions [17–19]. Fenton treatment is performed through four stages including pH adjustment, oxidation reaction, neutralization-coagulation and precipitation [20]. The Fenton process can be modified by electricity which is called electro-Fenton process [21,22]. The electro-Fenton process (FO and electro-coagulation combination) increases the degradability of organic compounds in a strong wastewater [23–25]. Fenton reagents can be added from out of reactor and inert electrodes with high catalytic activity are used while in the other configuration, hydrogen peroxide is added from out and Fe^{2+} ions are provided from a sacrificial cast iron anode [22].

In this research the reagents were added from out and iron electrodes as source of Fe^{2+} ions were used. Five independent variables including reaction time, current density, pH, H_2O_2/Fe^{2+} molar ratio and H_2O_2/Dairy wastewater (DW) (ml/l) were studied on COD and color removal from a dairy wastewater. Then, these parameters were optimized by a classical method [26]. Box-Behnken design and Response Surface Methodology (RSM) were used to design the experiments and optimize the operating conditions [27]. In fact,

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Table 1
Characteristics of the used wastewater.

Parameter	Unit	Value
Chemical oxygen demand (COD)	mg/l	2527
Color	Color unit	100
pH	–	6.27
Conductivity	μs/cm	210
TDS	mg/l	981

Table 2
Independent variables and their levels obtained from the statistical software.

Variable (unit)	Factors	Coded factors (X)		
	x_i	−1	0	+1
Reaction time (min)	x_1	10	50	90
Current density (mA/cm^2)	x_2	20	50	80
pH	x_3	3	7	11
H_2O_2/DW (ml/l)	x_4	0.3	1.22	2.14
Molar ratio	x_5	0.5	2.75	5

the RSM main benefit is number of experiments reduction and enough data preparation for the optimization.

2. Experimental

2.1. Materials and methods

2.1.1. Wastewater sampling and characterization

The present study was conducted on an industrial wastewater obtained from Malga Dairy Company (Arak, Iran). One sample was randomly taken from equalization basin and saved in a plastic vessel. Then, it was immediately transported to Arak University Chemical Engineering Research Laboratory and stored in a refrigerator at 4 °C before further analysis. The characteristics of this wastewater are reported in Table 1.

2.1.2. Electro-Fenton experiments

Electro-Fenton of dairy wastewater was carried out on the laboratory scale. This experimental apparatus has three main parts including DC power supply, digital magnetic mixer and 400 cm^3 beaker as reactor with two parallel plate ferrous electrodes (Fig. 1). Dimension of ferrous electrodes was 2 cm × 0.5 cm with an effective area of 1 cm^2. The electrodes gap was at 3 cm. pH of each sample was adjusted by H_2SO_4 or NaOH before the Fenton reagents addition. The pH was measured using a pH meter (METTLER-TOLEDO 320) which was calibrated with the standard buffers at room temperature. In each run, 250 cm^3 of wastewater was placed in baker and desired amounts of ferrous sulfate heptahydrate ($FeSO_4 \cdot 7H_2O$) and hydrogen peroxide (purchased from Merck with purity of 30%) were added to the cell before turning on the electricity. A direct current power supply (fabricated by Kala Gostaran-e-Farada supplier, 30 V and 3 A) was used to provide the desired electrical current. The effluent under treatment was homogenized by magnetic stirrer at 400 rpm. The agitation also allows separation of gases formed in the solution. It avoids foam formation which affects the batch process. All experiments were carried out at room temperature (25 °C ± 2 °C) and atmospheric pressure. DC power source was turned off and the reaction was stopped when the reaction time was found. Then, the samples were allowed to stay for 30 min (for solid sedimentation). After each experiment the electrodes were cleaned with distilled water to remove any sludge residues on the surfaces. COD and color were respectively measured at 228 nm and 346 nm wavelength using a UV–Vis spectrophotometer (HACH, US).

2.1.3. Experimental design

The Design Expert software (version 7.0.0) was used for the experimental design in this research. Five independent variables including reaction time (X_1), current density (X_2), pH (X_3), volumetric ratio of H_2O_2/DW ml/l (X_4) and H_2O_2/Fe^{2+} molar ratio (X_5) with three level full factorial Box-Behnken response surface experimental design (BBD) were used. Therefore, forty six experiments were designed. COD and color removal (Y_1 and Y_2) were considered as the dependent variables (response). According to the statistical software, five significant independent variables were converted to the dimensionless coded data for deactivating the units as [12]:

$$X_i = (x_i - x_0)/\Delta x \quad (1)$$

Where, X_i and x_i are the coded (without unit) and uncoded (with unit) variables, respectively. x_0 is x_i at the centre point and Δx is the difference between x_i and x_0. The variables were coded as −1, 0 and 1 for low, medium and high levels, respectively (Table 2).

The responses were explained as removal percentage. They are calculated using the following equation:

$$\text{Removal}(\%) = (C_i - C_0)/C_i \quad (2)$$

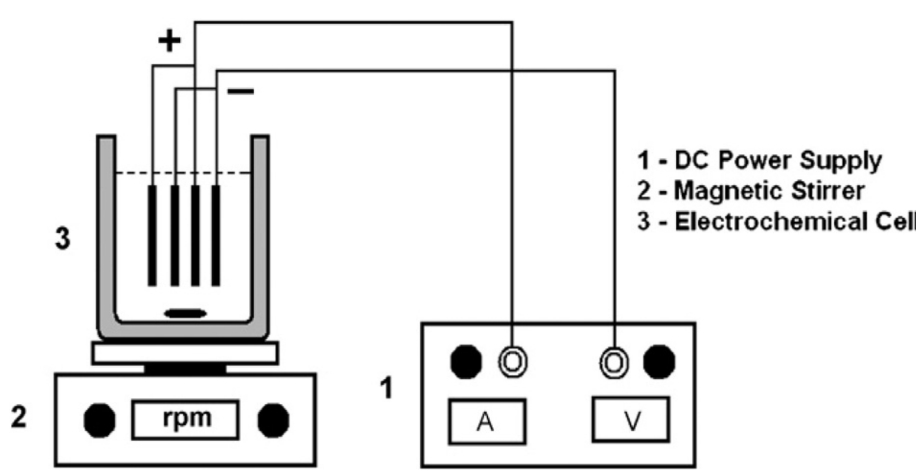

Fig. 1. Experimental appratus.