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Optimization of phosphate removal from wastewater by electrocoagulation with aluminum plate electrodes

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Abstract

The Taguchi method was used to determine the optimum conditions for the phosphate removal from wastewater by electrocoagulation with aluminum plate electrodes. The experimental parameters investigated were initial phosphate concentration, initial pH of the wastewater, supporting electrolyte concentration, supporting electrolyte type and current density. The ranges of experimental parameters were between 50 and 500 mg/L (as PO_4–P), 4–7 for initial pH, 0–10 mM, NaCl, $NaNO_3$, Na_2SO_4 and $CaCl_2$ and 0.25–1.00 mA/cm^2 mm for initial phosphate concentration, initial pH of the wastewater, supporting electrolyte concentration, supporting electrolyte type and current density, respectively. Reaction period was kept constant in 25 min for statistical analysis. The optimum conditions for these parameters were found to be 50 mg/L, 4, 5 mM, NaCl and 1.00 mA/cm^2, respectively. Under these conditions, the predicted and experimental removal efficiency of phosphate from wastewater by electrocoagulation with aluminum plate electrodes were 99.9 and 100.0%, respectively. A statistical analysis of variance (ANOVA) was performed to see whether the process parameters were statistically significant or not. According to the *F*-test results, it can be concluded that the degrees of the influences of parameters on the removal efficiency is initial phosphate concentration, current density and initial pH of the solution.
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Keywords: Taguchi method; Optimization; Phosphate removal; Electrocoagulation; Aluminum electrode

1. Introduction

As well known, eutrophication is one of the main problems nowadays encountered in the monitoring of the environmental water sources the industrialized countries. This phenomenon is caused by the excess phosphorus concentration in the effluents from municipal or industrial plants discharged in the environment [1] the usual forms of phosphorus found in solutions include orthophosphate, polyphosphate and organic phosphate [2]. The principal phosphorus compounds in wastewater are generally orthophosphate forms together with smaller amounts of organic phosphate [3]. In the countryside, where agriculture and animal husbandry are the main industries, wastes from these activities will contribute to the accumulation of P in soil and water bodies. These phosphorus compounds, dissolved in surface or ground waters, are responsible for the eutrophication in closed water systems, especially in lakes and enclosed bays where the water is almost stagnant [4]. Phosphorus removal techniques are chemical treatments like adsorption, chemical precipitation, ion exchange, electrodialysis, hybrid systems containing fly-ash adsorption and membrane filtration and electrocoagulation. Adsorption and chemical precipitation among the above methods have been widely used for phosphate removal [5–13]. The removal of phosphate from aqueous streams consists of the conversion of soluble phosphate to an insoluble solid phase. This solid phase can be separated from water by means of sedimentation or filtration. In wastewater applications, the most common and successful methods to precipitate phosphate involve the dissolved cations Al^{3+}, Ca^{2+}, Fe^{3+} and to a lesser extent of Fe^{2+}. It was found that when iron and aluminum are present in the water, $FePO_4$ and $AlPO_4$ forms in the low pH range (<6.5) and at higher pH range (>6.5) iron and aluminum increasingly convert to oxides and hydroxides. A higher pH is more ideal for precipitation of phosphate with calcium as apatites and hydroxyapatites [3].

In recent years, electrocoagulation has been successfully tested to treat wastewater. Electrocoagulation is a process consisting of creating metallic hydroxide flocks within the wastewater by electrodissolution of soluble anodes, usually made of iron or aluminum [14]. The difference between electro-

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coagulation and chemical coagulation is mainly in the way of aluminum ions are delivered. In electrocoagulation, coagulation and precipitation are not conducted by delivering chemicals – called coagulants – to the system, but via electrodes in the reactor [14]. Electrocoagulation is based on the fact that the stability of colloids, suspensions and emulsions is influenced by electric charges. Therefore, if additional electrical charges are supplied to the charged particles via appropriate electrodes, the surface charge of particles is neutralized and several particles combine into larger and separable agglomerates [15]. Electrode assembly is the heart of the treatment facility. Therefore, the appropriate selection of its materials is very important. The most common electrode materials for electrocoagulation are aluminum and iron. They are cheap, readily available, and proven effective [16]. When aluminum is used as electrode material, the reactions are as follows:

- At the cathode:

$$3H_2O + 3e^- \rightarrow \tfrac{3}{2}H_2(g) + 3OH^- \quad (1)$$

- At the anode:

$$Al \rightarrow Al^{3+} + 3e^- \quad (2)$$

- In the solution:

$$Al^{3+}(aq) + 3H_2O \rightarrow Al(OH)_3 + 3H^+(aq) \quad (3)$$

Taguchi's orthogonal array (OA) analysis is used to obtain the best parameters for the optimum process design with the least number of experiments. In recent years, the Taguchi method has been used to determine optimum parameters because of its many advantages [17]. The main advantages of this method over other statistical experimental design methods are that the parameters affecting an experiment can be investigated as controlling and not controlling and that the method can be applied to an experimental design involving a large number of design factors [18,19].

Aim of this study is not to investigate the treatability of the phosphate containing wastewater by electrocoagulation method. Our aim is to determine the optimum operating conditions such as initial phosphate concentration, supporting electrolyte type and concentration, current density and initial pH of the wastewater for the removal of phosphate from waters by electrocoagulation method with plate aluminum electrodes based on removal efficiency.

2. Experimental

2.1. Materials

All chemicals used were analytical grade and used without any further treatment. Distilled water was used in all experiments. Phosphate solutions were prepared from KH_2PO_4 (Riedel de Haën, 98%). NaCl (Merck, 99.5%), $NaNO_3$ (Merck, 99%), Na_2SO_4 (Sigma–Aldrich, 99%) and $CaCl_2$ (Merck, >90) were used as supporting electrolyte. Treated wastewater was collected over a desired period of time from the reactor and collected samples were filtered by the cellulose acetate membrane filter with the pore diameter of 045 μm (Schleicher and Schuell) before the analysis. The analysis of phosphate was carried out using the yellow vanadomolybdophosphoric acid method by a double beam spectrophotometer (Shimadzu UV-160 A) according to the Standard Methods for Examination of Water and Wastewater [20]. The initial pH was adjusted to a desired value using NaOH (Merck, 5N) or HNO_3 (Carlo Erbaa, 65%).

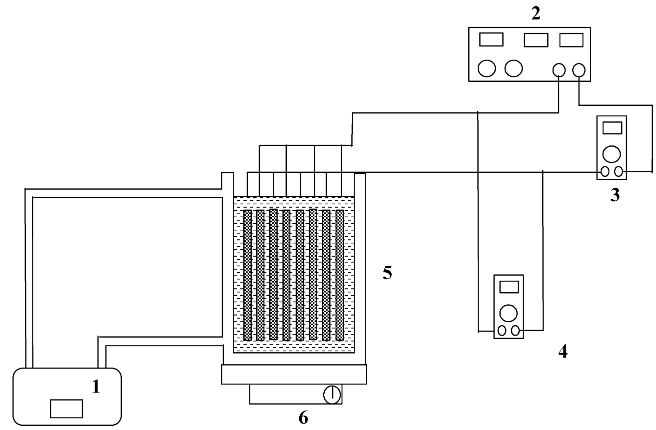

Fig. 1. Schematic diagram of the experimental setup.

2.2. Experimental setup and procedure

The experimental setup is schematically shown in Fig. 1. The electrocoagulation unit consists of six pair of electrodes made of plate aluminum with total area of approximately 1500 cm^2 and the gap between the electrodes is 5 mm. Electrodes were connected to a digital dc power supply (Shenzen-Mastech HY 3005-3) in monopolar mode. Two digital multimeters (Brymen Bm 201) as ampermeter and voltmeter were used to measure the current passing through the circuit and the applied potential, respectively. The electrocoagulation unit has been stirred at 150 rpm by a magnetic stirrer (Heidolp MR 3004 S). The experimental setup is shown in Fig. 1. The thermostated electrocoagulator is made of plexiglass with the volume of 850 mL. During the experiments, temperature, conductivity and pH of the wastewaters were measured by a multi-parameter (WTW Multiline P-4 F-Set-3). Reactor was operated in batch and galvanostatic mode.

2.3. Statistical analysis

The variables chosen for this investigation are supporting electrolyte type and concentration, current density, initial phosphate concentration, and initial pH of the wastewater. The variables investigated and their levels were summarized in Table 1. Reaction period was kept constant in 25 min for statistical analysis.

The use of the parameter design in the Taguchi method to optimize a process with multiple performance characteristics

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