Application of electrochemical peroxidation (ECP) process for waste-activated sludge stabilization and system optimization using response surface methodology (RSM)
Gagik Badalians Gholikandi and Khashayar Kazemirad

ABSTRACT
In this study, the performance of the electrochemical peroxidation (ECP) process for removing the volatile suspended solids (VSS) content of waste-activated sludge was evaluated. The Fe²⁺ ions required by the process were obtained directly from iron electrodes in the system. The performance of the ECP process was investigated in various operational conditions employing a laboratory-scale pilot setup and optimized by response surface methodology (RSM). According to the results, the ECP process showed its best performance when the pH value, current density, H₂O₂ concentration and the retention time were 3, 3.2 mA/cm², 1,535 mg/L and 240 min, respectively. In these conditions, the introduced Fe²⁺ concentration was approximately 500 (mg/L) and the VSS removal efficiency about 74%. Moreover, the results of the microbial characteristics of the raw and the stabilized sludge demonstrated that the ECP process is able to remove close to 99.9% of the coliforms in the raw sludge during the stabilization process. The energy consumption evaluation showed that the required energy of the ECP reactor (about 1.8–2.5 kWh (kg VSS removed)⁻¹) is considerably lower than for aerobic digestion, the conventional waste-activated sludge stabilization method (about 2–3 kWh (kg VSS removed)⁻¹). The RSM optimization process showed that the best operational conditions of the ECP process comply with the experimental results, and the actual and the predicted results are in good conformity with each other. This feature makes it possible to predict the introduced Fe²⁺ concentrations into the system and the VSS removal efficiency of the process precisely.

Key words | coliforms removal, electrochemical peroxidation (ECP) process, energy consumption, response surface methodology (RSM), VSS removal efficiency, waste-activated sludge stabilization

INTRODUCTION
Sludge management, especially waste-activated sludge stabilization, is a fundamental area of concern in conventional wastewater treatment plants. Waste-activated sludge, which is considered to be a clear potential threat for the environment, is the major by-product of the wastewater treatment process (Bernal-Martínez et al. 2010; Ghatak 2014). There is therefore a demand for novel treatment technologies that are capable of achieving significant removal efficiencies with more convenient operational conditions. Recently, the electrochemical advanced oxidation processes (EAOPs) have been considered for wastewater treatment (Babuponnusami & Muthukumar 2014). The efficiency of advanced oxidation processes (AOPs) in the degradation of biological organic compounds, and toxic and chemically resistant substances, has made these processes superior to research or use (Oturan & Aaron 2014; Wang et al. 2016). Traditionally, it is considered that AOPs include a large variety of degradation processes in which very oxidizing and unstable species have the predominant role. Among these radicals, the hydroxyl radical is considered the most important free radical in chemistry and biology due to its multiple implications and applications (Brillas et al. 2009). This radical is one of the most impressive oxidizing agents (E₀ = 2.87) and reacts non-selectively to remove the
related reactions are represented by Equations (1) and (2).

$$\text{Fe} \rightarrow \text{Fe}^{2+} + 2e^- \quad (1)$$

$$\text{Fe}^{2+} + \text{H}_2\text{O}_2 \rightarrow \text{Fe}^{3+} + \text{OH}^0 + \text{OH}^- \quad (2)$$

Several studies carried out on the ECP process have focused on water treatment. Due to the ability of the ECP process to remove various contaminants, it could be also appropriate for waste-activated sludge stabilization (Chiarenzelli et al. 2001; Gutiérrez et al. 2010; Kokkali 2011; Liu et al. 2012; Ozyonar & Karagozoglu 2015). However, there is a lack of assessment of iron electrode functionality, as one of the most influential contributing factors in the ECP process.

With this in mind, we investigated the effectiveness of this technology for waste-activated sludge stabilization at a laboratory scale. The main objective of the present study was to consider the effects of various operational parameters on introduced Fe$^{2+}$ concentrations and volatile suspended solids (VSS) removal efficiency as the important factors of the ECP process.

Due to the remarkable dependency of the ECP process on pH, H$_2$O$_2$ concentration, current density and retention time (Umar et al. 2010; Nidheesh & Gandhi 2012; Sirés et al. 2014), these factors were considered as input variables, and the introduced iron ions (Fe$^{2+}$) and the VSS removal efficiency as responses. In order to optimize the performance of the ECP process in this study, response surface methodology (RSM) was used to develop a mathematical correlation between the operational parameters of the ECP process and its responses. This methodology is a successfully applied statistics-based method for designing experiments, evaluating the individual and interaction effects of independent variables, and optimizing the process parameters with a limited number of experiments (Ghafari et al. 2009; Zhu et al. 2011; Rasouli Sadabad & Gholikandi 2017). In addition, given the significance of the electrical energy consumption of the ECP reactor and the microbial characteristics of stabilized sludge, these two factors were also investigated in the optimum operating conditions.

### MATERIALS AND METHODS

#### Materials

The pilot studies were conducted at the laboratory scale (Figure 1). The waste-activated sludge studied was obtained from Shahid Mahalati urban sewage treatment plant which is located in northeastern Tehran, Iran. The characteristics of this waste-activated sludge are presented in Table 1.

As shown in Figure 1, the reactor was made up of a one litre Plexiglas cylinder, containing 0.9 litres of the waste-activated sludge sample, fitted with four iron electrodes (purity >99.5%). The electrode dimensions were 140 mm × 60 mm × 1 mm, and were made from iron in order to supply the Fe$^{2+}$ required as a catalyst for to the reaction shown in Equation (2). In order to study the rate of the introduced Fe$^{2+}$ to the system via the electrodes, an inductively coupled plasma spectrometer (ICP; 7500; P/N 211-46500-92) measuring apparatus was used to measure the introduced Fe$^{2+}$ concentration. The insertion depth (inside the sludge) and the aspect ratio of each electrode with sludge were about 100 mm and 100 mm × 60 mm, respectively. Also, the distance between the electrodes was 1.5 cm (Nidheesh & Gandhi 2012). An electric motor (Zheng, zs-r, 6(V) DC, 366 rpm) was used to provide proper mixing in the system. To set the current and voltage consumption of the system, a digital power supply (DAZHENG, DC Power, ps-305d, 0–5 A, 0–30 V) was used and polarity reversal was used for changing the polarity of the electrodes.
All the chemicals used in this study were obtained from Merck group, Germany. Sodium hydroxide (50% purity, 0.1 N) and sulphuric acid (98% purity, 0.1 N) were used in the experiments for pH adjustment. The lauryl tryptose broth (LTB), brilliant green bile (BGB), *Escherichia coli* (EC) and plate count agar (PCA) cultures were used to study and measure the pre- and post-experiment values of the following sludge microbial characteristics: coliform, total coliform, heat-resistant coliforms, and heterotroph bacteria.

### Sampling and test methods

First, the rate of Fe$^{2+}$ introduced into the system was studied in order to obtain a thorough understanding of the electrodes’ performance under different operational conditions and the effect of operational parameters on the electrodes. Current density (mA/cm$^2$), retention time (min), pH and H$_2$O$_2$ concentration (mg/L) were considered as the operational parameters and introduced Fe$^{2+}$ concentrations and the VSS removal efficiencies as the responses of the ECP process. The current polarity was reversed every 3 min in order to avoid passivation and to reach equal dissolution of both electrodes. During this stage, hydrogen peroxide was injected into the system and the sludge sample pH was adjusted by adding sulphuric acid or NaOH. Upon connection to the power supply, the electrodes were placed into the system and the voltage consumption was measured after adjusting the current to the desired level. After the test, sampling was performed at a sludge depth of 100 mm and the concentration of the Fe$^{2+}$ determined using the ICP apparatus. Upon completion of the test, the VSS removal efficiency, final voltage and microbial characteristics of the outlet sludge were measured. All the tests were conducted in conformity with the standard method (APHA, AWWA & WEF 2012). Each group of tests was repeated three times.

### Experimental design and data analysis

The results of the experiments were optimized by Design-Expert® software (Stat-Ease Inc., Minneapolis, USA) using central composite design (CCD) analysis. The experiments were designed through the software and carried out in the conditions determined. Then the results of the experiments

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**Table 1** | Excess sludge specification

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>6.53–7.31</td>
</tr>
<tr>
<td>Temperature(ºC)</td>
<td>13.5–15.7</td>
</tr>
<tr>
<td>COD (mg/L)</td>
<td>5,768–10,985</td>
</tr>
<tr>
<td>VSS (mg/L)</td>
<td>3,136–6,682 (2.6–5.6 (g/kg))</td>
</tr>
<tr>
<td>VS (mg/L)</td>
<td>3,201–6,810 (2.7–5.8 (g/kg))</td>
</tr>
<tr>
<td>TS (mg/L)</td>
<td>4,375–8,825 (3.8–7.6 (g/kg))</td>
</tr>
<tr>
<td>TSS (mg/L)</td>
<td>4,085–8,537 (3.5–7.4 (g/kg))</td>
</tr>
<tr>
<td>VSS/TSS</td>
<td>0.76–0.78</td>
</tr>
</tbody>
</table>

COD – chemical oxygen demand; VS – volatile solids; TS – total dry solids; TSS – total suspended solids.
were inserted to the model in order to identify the coefficients and effects of each parameter.

RESULTS AND DISCUSSION

Effect of independent parameters on the introduced Fe$^{2+}$ and the VSS removal efficiency

Iron is recognized as the best catalyst for the Fenton-based processes (Sirés et al. 2014). Controlling the amount of Fe$^{2+}$ obtained from iron electrodes in the ECP process would not only limit and/or inhibit negative effects on the reaction, represented by Equation (3), and the available Fe$^{2+}$ concentration in the system, but also increase the efficiency of the studied system (Abdessalem et al. 2008; Brillas et al. 2009).

$$\text{Fe}^{2+} + \text{OH}^0 \rightarrow \text{Fe}^{3+} + \text{OH}^- \quad (3)$$

Due to the important role of the Fe$^{2+}$ introduced into the system from the electrodes on the effectiveness of the ECP process, we first studied the effect of each individual parameter on this response, and then the effect of the mentioned parameters and introduced Fe$^{2+}$ on the extent of the VSS removal efficiency.

pH

The catalytic performance of the introduced Fe$^{2+}$ concentration strongly is dependent on the pH value of the solution (Duesterberg et al. 2008). In order to evaluate the effect of pH, the other operational parameters were kept approximately constant ($\text{H}_2\text{O}_2 = 1.695 \text{ mg/L}$, current density = 2.8 mA/cm$^2$, retention time = 120 min), and the experiments were carried out at various pH values. Figure 2 shows the experimental results of the introduced Fe$^{2+}$ and the VSS removal efficiency in the different operating conditions. Also, Figure 2(a) shows the VSS removal efficiencies and the introduced Fe$^{2+}$ concentrations in different pH values of the process. According to Figure 2(a), the introduced Fe$^{2+}$ concentration in the system from the electrodes was dependent on pH values as well as the VSS removal efficiency. The introduced Fe$^{2+}$ concentration did not show significant change at pH values lower than 5, but increasing the pH value to 6 led to a sudden reduction. Further increasing the pH to values greater than 6 showed an increasing trend with a slight slope, which is negligible. Also, according to Figure 2(a), the maximum VSS removal efficiency was 67% at pH = 3. Moreover, for pH values lower than 2 and higher than 5, the VSS removal efficiency decreased considerably. Based on these results and observations, it seems that at pH values higher than 5, the Fe$^{3+}$ species would be deposited in the form of Fe(OH)$_3$, which leads to the reduction of the formation and introduction of hydroxyl radicals into the system as well as the spontaneous decomposition of hydrogen peroxide (Shemer & Linden 2006). At pH values lower than 2, Fe-containing complexes such as [Fe(H$_2$O)$_6$]$^{2+}$ which react much more slowly with hydrogen peroxide than other species are able to be formed (Wang et al. 2010). Moreover, in the presence of high H$^+$ concentrations, hydrogen peroxide would decompose and be converted into the stable species ([H$_2$O$_2$]$^+$) which reacts with Fe$^{2+}$ slower than hydrogen peroxide (Zhou et al. 2007; Ghoneim et al. 2011).

Current density

The current density is an important functional parameter in the Fenton-based reaction processes since it affects subsequent H$_2$O$_2$ production, Fe$^{2+}$ reduction and OH$^0$ production (Zhang et al. 2007; Sirés et al. 2014). In order to evaluate the effect of the current density, the other operational parameters were kept approximately constant ($\text{H}_2\text{O}_2 = 1.695 \text{ mg/L}$, $\text{pH} = 3$, retention time = 120 min), and the experiments were carried out at various current densities. Figure 2(b) shows the introduced Fe$^{2+}$ concentrations and the VSS removal efficiencies at various current densities. As shown in Figure 2(b), in the absence of current, no Fe$^{2+}$ is produced from the electrodes and the VSS removal reactions do not happen. Increasing the current density leads to an almost linear increase in the introduced Fe$^{2+}$ concentration. Therefore, the behaviour of iron electrodes can clearly be observed at the different current densities. Based on the results, increasing the current density increases the efficiency of the system, particularly OH$^0$ generation (Sirés et al. 2014).

According to Figure 2(b), the maximum VSS removal efficiency of the process (70%) was obtained at the current density of 5.2 mA/cm$^2$. Further increasing the current density (from 3.2 to 4.17 mA/cm$^2$) led to reducing the VSS removal efficiency to 64%. Thus, an increase in current density further increased undesired reactions and reduced the VSS removal efficiency of the system (Brillas et al. 2009; Sirés et al. 2014).
**H₂O₂ concentration and [H₂O₂]/[Fe²⁺] ratio**

In order to evaluate the effect of H₂O₂ concentration and [H₂O₂]/[Fe²⁺] ratio, the other operational parameters were kept approximately constant (pH = 3, current density = 3.2 mA/cm², retention time = 120 min), and the experiments were carried out at various H₂O₂ concentrations and [H₂O₂]/[Fe²⁺] ratios. Figure 2(c) and 2(e) represent the results of evaluating these parameters. As shown in Figure 2(c) the presence or absence of hydrogen peroxide has no significant effect on the introduced Fe²⁺ concentration, but change the VSS removal efficiency remarkably. In
the absence of H₂O₂, the VSS removal efficiency of the process was zero. The VSS removal efficiency shows an upward trend by increasing the H₂O₂ concentration to 1,535 mg/L (the ratio [H₂O₂]/[Fe²⁺] = 6.28, shown in Figure 2(e)), but at higher concentrations, this upward trend reverses. The maximum VSS removal efficiency for the sludge was obtained as 71% in these conditions. In other words, the system efficiency decreased as a result of Equation (4) which occurred within the 6.28 < [H₂O₂]/[Fe²⁺] < 7.92 range. It seems that at higher concentrations of H₂O₂, the system efficiency decreased by consumption of hydroxyl radicals in reaction, represented by Equation (4), and the subsequent reduction in the concentration of hydroxyl oxidizing agents (Muruganandham & Swaminathan 2004).

\[ OH^\bullet + H_2O_2 \rightarrow HO_2^\bullet + H_2O \] (4)

Retention time

Because of the long retention time demanded by most conventional sludge stabilization methods (aerobic/anaerobic digestion), one of their significant disadvantages, the much shorter retention time of EAOPs is considered to be a noticeable advantage (Huang et al. 2001; Gholikandi et al. 2014; Oturan & Aaron 2014). Accordingly, the required retention time in the ECP process for stabilization of the waste-activated sludge was considered one of the operational parameters in the current study. In order to evaluate the retention time, the values of the other operational parameters were kept approximately constant (pH = 3, current density = 3.2 mA/cm², H₂O₂ = 1,535 mg/L), and the experiments were carried out using various retention times. Figure 2(d) shows the introduced Fe²⁺ concentrations and the VSS removal efficiencies in various retention times. As shown in Figure 2(d), increasing the retention time increases (approximately linearly) the introduced Fe²⁺ concentration into the system, until it reached 780 mg/L, in 6 hours. Also, in 2 and 4 hours after the start of the test, the sludge VSS removal efficiency had reached 71% and 74%, respectively. Because the efficiency exhibited no significant change after 4 hours of the test, it was concluded that the optimum retention time and the maximum VSS removal efficiency in the ECP system were 4 hours and 74%, respectively.

Altering the microbial characteristics of the waste-activated sludge

The results obtained from evaluating the microbial characteristics of the raw and stabilized sludge by the ECP process in the laboratory demonstrated that this system had a highly acceptable performance (99.9% efficiency) in altering the microbial characteristics of the sludge. Table 2 shows the microbial characteristics of raw and stabilized waste-activated sludge. Considering the high degree of coliform removal, the stabilized waste-activated sludge was placed in Class A of the US EPA standard. The status of the microbial characteristics makes this sludge able to be reused, particularly in agriculture.

Electrical energy consumption

The electrical energy consumption of the system was calculated based on the voltage consumption data obtained from the supply source connected to the electrodes at the optimum operational conditions of the laboratory-scale setup. In order to reduce possible errors, the experiments were repeated three times and the electricity consumption was calculated. Electricity consumption of the system during the experiment was calculated using Equation (5) (Ghosh et al. 2011).

\[ \text{UED} = \frac{(V \times I \times t)/v}{(\text{VSS}_{\text{in}}/1000) \times (\eta/100)} \] (5)

where UED (kWh per kg of the removed VSS) is considered as the required electrical energy, V as the consumed voltage (V), I as the applied current (A), t as the test retention time (h), v as the system volume (L), VSS_in as the inlet sludge rate (mg/L), and η as the VSS removal efficiency (expressed as a percentage) of the system.

<table>
<thead>
<tr>
<th>Table 2: Raw and stabilized sludge microbial characteristics</th>
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<tr>
<td></td>
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<tr>
<td>---------------------------</td>
</tr>
<tr>
<td>Raw sludge</td>
</tr>
<tr>
<td>Stabilized sludge</td>
</tr>
<tr>
<td>Growth medium</td>
</tr>
<tr>
<td>Removal efficiency (%)</td>
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</table>
Thus, the energy consumption was obtained as 1.8–2.5 kWh per kg of removed VSS. In contrast, based on the available resources, the VSS removal in the conventional bio-aerobic method used for stabilization of the waste-activated sludge (Wang et al. 2008) is reported between 35% and 50% with the corresponding energy consumption of 2–3 kWh per kg of VSS removal (Tchobanoglous 1979). Therefore, as compared with aerobic digestion, as a conventional system, the ECP reactor provided approximately twice the efficiency, and less electricity consumption.

**Response surface methodology**

**Statistical analysis**

RSM is considered a useful statistical technique for designing, modeling, analyzing, and optimizing water and wastewater chemical reactions (Benatti et al. 2006; Bashir et al. 2010). In this study, CCD, as the most widely applied technique in RSM, was utilized in order to optimize the introduced Fe\(^{2+}\) concentration and the VSS removal efficiency. Statistical analysis of the model was carried out to evaluate the analysis of variance (ANOVA) using Design-Expert® 7.0.0 software. The pH value, current density (mA/cm\(^2\)), \(\text{H}_2\text{O}_2\) (mg/L) and retention time (min) were selected as the four main experimental factors in order to evaluate the effect of operational parameters (variable factors) on the responses. Also, it is possible to represent the independent operational parameters and the response in quantitative forms:

\[
y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_i x_i^2 + \sum_{i<j}^{k} \beta_{ij} x_i x_j + e_i \tag{6}
\]

where \(y\) is the response, \(x_i\) and \(x_j\) are variables, \(\beta_0\) is constant coefficient, \(\beta_i, \beta_i\) and \(\beta_{ij}\) are the interaction coefficients of the linear, quadratic and the second-order terms, respectively, and finally \(e_i\) is the error. In this study, the introduced Fe\(^{2+}\) concentration and the VSS removal efficiency data were processed for Equation (6) including ANOVA to obtain the interaction between the process variables and the responses.

**Determination of the effective parameters range and experimental design**

Considering the theoretical studies and the results of laboratory experiments, a CCD was carried out using selected minimum and maximum levels. The evaluated ranges used in the study were 0–2,000 mg/L for \(\text{H}_2\text{O}_2\) concentrations, 0–360 min for retention time, 1–9 for initial pH and 0–4.17 mA/cm\(^2\) for current density. Table 3 shows the factors’ coded values which were set at five levels: –2 (minimum), –1, 0 (central), +1 and +2 (maximum). The responses are shown as the VSS removal efficiency (%) and the introduced Fe\(^{2+}\) concentration (mg/L).

Table 4 shows the results of CCD experiments for evaluating the effect of four independent variables along with the predicted mean and observed experimental results, shown as the VSS removal efficiency and the introduced Fe\(^{2+}\) concentration. The optimum values of the chosen variables were obtained by solving the regression equation and analyzing the response surface contour plots (Noordin et al. 2004). The change independent variables were explained by the multiple coefficients of determination, R\(^2\). The model equation was utilized in order to predict the optimum value and subsequently, elucidate the interaction between the factors in the specified range (Elibol 2002). Thirty experiments were conducted in duplicate according to the scheme mentioned in Table 4.

**Analysis of variance**

ANOVA was employed to analyze the effect of different factors on the introduced Fe\(^{2+}\) concentration in the system and the VSS removal efficiency. ANOVA results of the quadratic models of the introduced Fe\(^{2+}\) concentration and the VSS removal efficiency are shown in Table 5. The lower \(p\)-values (or higher F-values) imply that the addition of higher-order terms might improve the model (Hamsaveni et al. 2001; Noordin et al. 2004; Singh et al. 2011). The ANOVA results showed that two-factor interactions (2FI) and the quadratic model were appropriate enough for the introduced Fe\(^{2+}\) concentration and the VSS removal efficiency, respectively. The high model F-values (42.96 and

**Table 3 | Experimental range and levels of independent variables**

<table>
<thead>
<tr>
<th>Factors</th>
<th>Range and levels (coded)</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>(A) 1 3 5 7 9</td>
</tr>
<tr>
<td>Current density (mA/cm(^2))</td>
<td>(B) 0 1.04 2.09 5.13 4.17</td>
</tr>
<tr>
<td>(\text{H}_2\text{O}_2) (mg/L)</td>
<td>(C) 0 500 1,000 1,500 2,000</td>
</tr>
<tr>
<td>Retention time (min)</td>
<td>(D) 0 90 180 270 360</td>
</tr>
</tbody>
</table>
The estimated coefficients of functions were obtained by modifying the models. They are given in Equation (7) for the introduced Fe$^{2+}$ concentration, and in Equation (8) for the VSS removal.

For the introduced Fe$^{2+}$ concentration:

$$y = +409.89 + 26.61 A + 156.67 B + 7.37 C$$
$$+ 213.79 D - 52.12 AB + 1.34 AC + 23.43 AD$$
$$+ 1.51 BC + 70.20 BD + 1.82 CD$$

(7)

For the VSS removal:

$$y = +142.55 - 33.44 A + 42.90 B + 24.92 C + 21.82 D$$
$$- 31.54 AB - 1.57 AC - 5.45 AD + 11.43 BC$$
$$+ 7.95 BD + 2.72 CD - 5.73 A^2 - 14.70 B^2$$
$$- 17.44 C^2 - 15.25 D^2$$

(8)

where A, B, C and D are the four independent variables. The significance of each coefficient present in Equations (7) and (8) was determined by the student’s t-test and p-values (Noordin et al. 2004).

Diagnostic plots such as the predicted versus actual values (Figure 3) are useful in order to judge the model satisfaction. The predicted versus actual values plots of parameters are presented in Figure 3(a) and 3(b) for the introduced Fe$^{2+}$ concentration and the VSS removal.
In these figures the values of $R^2$ and $R_{adj}^2$ were evaluated as 0.9598 and 0.9387 for the introduced $\text{Fe}^{2+}$ concentration, and 0.9573 and 0.9174 for the VSS removal efficiency. These plots demonstrate an adequate agreement between real data and the ones obtained from the models.

The results of Design-Expert®

The effect of the operating parameters of pH, current density, $\text{H}_2\text{O}_2$ concentration and retention time on the introduced $\text{Fe}^{2+}$ concentration and the VSS removal efficiency are shown in Figure 4. The obtained results (Figure 4(a)), show that the introduced $\text{Fe}^{2+}$ concentration undergo no significant change in the $3 < \text{pH} < 4$ range, but in the $4 < \text{pH} < 7$ range demonstrate a reduction with a significant slope. Further increase in pH values over 5 did not have a noticeable effect on the introduced $\text{Fe}^{2+}$ concentration. As shown in the Figure 4(a), increasing current density would lead to an almost linear increase in the introduced $\text{Fe}^{2+}$ concentration. Also, according to Figure 4(b), increasing retention time would lead to an almost linear increase, while increasing $\text{H}_2\text{O}_2$ concentration will not have a considerable effect on the introduced $\text{Fe}^{2+}$ concentration. In addition, Figure 4(a) and 4(b) shows that the introduced $\text{Fe}^{2+}$ concentration dependency on the retention time and the current density is higher than pH and $\text{H}_2\text{O}_2$ concentration.

As shown in Figure 4(c) and 4(d), the VSS removal efficiency shows an upward trend with an increase in the current density, the retention time and $\text{H}_2\text{O}_2$ concentration up to about 3.13 mA/cm$^2$, 1,500 mg/L and 180 min, respectively. However, increasing any of these parameters further not only shows no effect on this response but also shows a decreasing trend. In addition, this response shows an increase with pH values up to 3, but at pH values higher than 3 (up to 7) show a decrease from 75% to 31%. Therefore, optimum values of the study parameters were identified for optimum VSS removal efficiency. According to Figure 4, in the optimum operating conditions of the ECP process (pH = 3, current density = 3.13 mA/cm$^2$, retention time = 240 min, and $\text{H}_2\text{O}_2$ = 1,500 mg/L), the VSS removal efficiency would be 74%.

CONCLUSION

The performance of the ECP process was evaluated in order to remove the VSS content of the waste-activated sludge, in various operational conditions and optimized by RSM. The introduced $\text{Fe}^{2+}$ concentration and the VSS removal efficiencies were considered as parameters that represent the ECP process performance at various pH values, current densities, $\text{H}_2\text{O}_2$ concentrations and retention times (as operational parameters), and the resulting values inserted into the model in order to be optimized by RSM. The results showed that the ECP process was able to obtain approximately 500 mg/L introduced $\text{Fe}^{2+}$ concentration and remove 74% of the VSS content of the waste-activated sludge at its best performance operational conditions (pH = 3, current density = 3.2 mA/cm$^2$, $\text{H}_2\text{O}_2$ = 1,535 mg/L, retention time = 240 min). Also, the energy consumption
evaluations showed that the required energy of the ECP reactor is about 1.8–2.5 kWh (kg VSS removed)\(^{-1}\) which is considerably lower than aerobic digestion, a conventional waste-activated sludge stabilization method (about 2–3 kWh (kg VSS removed)\(^{-1}\)). The RSM optimization process showed that the best operational conditions of the ECP process agrees with the experimental results. This feature makes it possible to predict the introduced Fe\(^{2+}\) concentrations into the system and the VSS removal efficiency of the process precisely. The results regarding the microbial characteristics of the raw and the stabilized sludge showed that the ECP process is able to remove about 99.9% of the coliforms in the raw sludge during the stabilization process. According to the results, it can be concluded that the ECP is an absolutely environmentally friendly process which is not only able to remove twice the VSS of waste-activated sludge as aerobic digestion in a noticeably shorter retention time, but also consumes noticeably less electrical energy.

**REFERENCES**


Kokkali, V. 2011 Electrochemical peroxidation of contaminated water and assessment of the toxicity using existing and novel bioassays.


Rasouli Sadabad, H. & Badalians Gholikandi, G. 2017 Harvesting direct electricity from municipal waste-activated sludge simultaneous with its aerobic stabilization process:


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