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Enhanced electrocoagulation–electroflotation for turbidity removal by *Opuntia ficus indica* cladode mucilage

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Abstract

The optimisation of the electrocoagulation-electroflotation (EC-EF) process assisted by the mucilage of the *Opuntia ficus indica* (OFI), on the turbidity removal was performed through the response surface methodology (RSM). For a solution of 300 mg/L of silica gel, high turbidity removal (93.14% $\pm$ 1.31) was obtained under the optimal conditions of 2.5 mg/L, 21.2 V, 9.65 and 2.61 mS/cm for the mucilage concentration, voltage, pH and conductivity, respectively, this experimental value was close to the predicted value of 92.96% $\pm$ 0.3. OFI mucilage increases turbidity removal efficiency and reduces specific energy consumption at a fixed current density. The turbidity removal of the EC-EF process was improved by 30.94% compared with the conventional EC–EF (EC–EF without OFI mucilage) which shows 62.02% $\pm$ 1.45 of turbidity removal.

Introduction

For many years, scientific community has been testing new methods for water treatment. Some processes are rather well known, such as filtration, disinfection or coagulation, but new cheaper and affordable water treatment processes are needed (Antov *et al*. 2012; Alila & Boufi 2009). Namely, coagulation/flocculation step, which is an essential process in the treatment of both surface water and industrial wastewaters, includes removal of dissolved organic species and turbidity from water most commonly via addition of conventional chemical based coagulants: aluminium, ferric chloride and synthetic organic polymers. While there are disadvantages associated with their usage such as ineffectiveness in low-temperature water (Haarhoff & Cleasby 1988), relatively high procurement costs, detrimental effects on human health, production of large sludge volumes. Electrocoagulation–electroflotation (EC–EF) was used for various wastewater treatment because of its *in situ* generation of coagulating agents by electro-oxidation of the anodes (Jiang *et al*. 2002; Chen *et al*. 2000; Esfandyari *et al*. 2015). The addition of natural coagulants may reinforce the efficacy of the EC–EF treatment. It was found that the EC–EF was more efficient for turbidity removal when assisted with OFI pad juice than the juice-free process (Adjeroud *et al*. 2015). Similarly, Zhang *et al*. (2006) found that the association of the cactus coagulant with aluminium chloride results in a better efficacy of the conventional coagulation-flocculation sewage treatment then the aluminium chloride alone.

EC–EF produces coagulants *in situ* by electrical dissolution of aluminium. After electrolysis, the cathode releases the hydrogen gas that aid to fleet the flocculants particles out of the water, these flocculants formed by the oxidation of the anodes that generate the metal ions responsible of destabilisation and aggregation of the suspend particles. Flocculants are formed by the oxidation of the anodes, that generate metal ions responsible of destabilisation and aggregation of the suspend particles. After electrolysis, the cathode releases hydrogen gas that aid to fleet the flocculants particles of the water (Chen *et al*. 2000; Chen 2004; Kobya *et al*. 2006b; Adjeroud *et al*. 2015). In the case of aluminium, main reactions are as follows:

$$\text{Anode}: \quad Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^- \qquad (1)$$

$$\text{Cathode} \qquad 3H_2O_{(1)} + 3e^- \rightarrow \frac{3}{2}H_{2(g)} + 3OH^-_{(aq)} \qquad (2)$$

After that, monomeric and polymeric aluminium species are generated following the interaction of Al^{3+} and OH^- which transform finally into $Al(OH)_{3(s)}$ according to the following equation:

$$Al^{3+} + 3H_2O_{(1)} \rightarrow Al(OH)_{3(s)} + 3H^+_{(aq)} \qquad (3)$$

The use of natural coagulant for wastewater treatment could be an option with many advantages over chemical agents, particularly the biodegradability, low toxicity and low residual sludge production (Narasiah *et al*. 2002). These advantages are especially amplified mainly that *Opuntia ficus indica* (OFI) is largely widespread, grows easily with very few water requirements in the local community. In Algeria, numerous efforts are being made for the exploitation of this plant. According to the HCDS (haut commissariat pour le développement de la steppe) the surface area of the cultivated or wildly grown plants is estimated to 31 359 ha in the region of Hodna, and 24 322 ha in the east region in 2015. A Nopal fruit crop can generate production of 20 tons per hectare annually and produce up to 50 tons of dry material with the potential use as a natural coagulant. In addition, the cost of the extraction process is low. In recent numerous studies, a variety of plant materials have been reported as a source of natural coagulants (Miller *et al*. 2008; Šćiban *et al*. 2009; Ramavandi 2014; Ramavandi *et al*., 2015). Recently, there are many information in the literature evaluating and exploiting the advantageous use of OFI, usually known as Nopal or prickly pear, in water treatment (Miller *et al*. 2008; Torres & Carpinteyro-Urban 2012; Barka *et al*. 2013; Bouatay & Mhenni 2014; Betatache *et al*. 2014; Fedala *et al*. 2015a). The cactus produces a gum-like substance; mucilage, which shows excellent flocculating abilities and is an economically viable alternative for low-income communities (Miller *et al*. 2008). The mucilage is a polysaccharide containing a molecule structure up to 30 000 monosaccharides (Felkai-Haddache *et al*. 2015) composed basically of arabinose, galactose, rhamnose, xylose and galacturonic acid (Majdoub *et al*. 2001; Felkai-Haddache *et al*. 2015). It helps cactus to retain water (Barbera *et al*. 1995).

In the literature, the efficacy of EC–EF treatment can be enhanced with additional synthetic (Can *et al*. 2006) or natural coagulants (Seid-Mohammadi *et al*. 2014; Adjeroud *et al*. 2015). Thus, in a green technological context, OFI juice was used as a natural coagulant/flocculant in EC–EF turbidity removal (Adjeroud *et al*. 2015), but this is the first study about enhancement of EC–EF turbidity water treatment using OFI mucilage, to which the role of coagulation is attributed. The objectives of this work are to examine the effect of the OFI cladode mucilage on the enhancement of the EC-EF turbidity removal and to optimise some parameters (pH, voltage, conductivity and mucilage concentration) by response surface methodology (RSM) to maximise EC–EF efficiency.

Materials and methods

Plant material

OFI cladodes without thorns were harvested from Aokas (Bejaia) located in North-East of Algeria in January 2015. The harvested cladodes were washed with tap water to remove all impurities, and then dried at room temperature during 30 min. The cladodes were cut in small pieces, crushed in a domestic blender to obtain the pulp that will serves to the mucilage extraction.

Mucilage extraction

The crushed Nopal cladode was homogenised with distilled water in the ratio of 1 : 4 (v/v) during maceration time of 4 h and at room temperature. The extract was filtered through a fine cloth. The mucilage was precipitated with ethanol (95%, v/v) in the ratio of 1 : 3 (v/v). The precipitate was washed three times with ethanol, then it was dried for solvent evaporation in a ventilated oven for approximately 1 h at 40°C, before freezing (−20°C) and lyophilisation at −50°C for 24 h.

EC–EF batch unit

The experimental set-up is schematically shown in Fig. 1. The EC–EF cell had two aluminium electrodes, each with a dimension of (27 mm × 17 mm × 1 mm). The electrodes were placed vertically and with a net spacing of 1 cm in the centre of the reactor. The electrodes were connected to a DC power supply (Statron Typ 3217, Bielefeld, Germany) providing 0–30 V (0–10 A), the electric current was conduct continuously to maintain a consistent influent to the treatment system. At every handling, 2 L of the turbid water was treated at room temperature.

Analysis

Turbidity measurement

The turbidity of the water was created by dissolution of an amount of 300 mg of silica gel (Woelm Pharma) in 1000 mL of tap water. A volume of 2000 mL of wastewater was used (Merzouk *et al*. 2009; Adjeroud *et al*. 2015). Samples turbidity was determined by UV–VIS spectrophotometer (Spectro Scan 50, Tokyo, Japan). Silica gel concentration was estimated from its absorbance characteristics at maximum wavelength (λ_{max}) of 740 nm (Merzouk *et al*. 2009; Adjeroud *et al*. 2015). The calculation of turbidity removal efficiency (TR) after electrochemical mucilage free treatment or

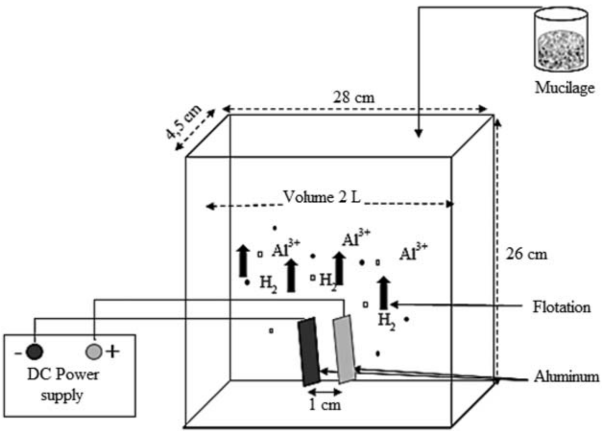

Fig. 1. Batch electrocoagulation–electroflotation unit assisted by the mucilage of *Opuntia ficus indica*.

mucilage assisted-treatment was performed using the following formula:

$$TR(\%)=\frac{C_0-C}{C_0}.100 \quad (4)$$

where C_0 and C are concentrations of silica gel before and after EC–EF process in mg/L, respectively.

pH and conductivity measurements

The pH of samples was measured with a pH-meter (pH 211; HANNA Instruments, Padova, Italy). The desired pH was attained by addition of appropriate volumes of HCl (1 M) or NaOH (1M). Conductivity was measured with multiparameter (Extech Instruments, Paris, France). The desired conductivity was adjusted by adding appropriate volumes of (1 M) NaCl.

Specific electrical energy consumption

The specific electrical energy consumption (SEEC) per kg turbidity (T) removed (E_T) under steady-state conditions was calculated as follows:

$$E_T=\left(\frac{\text{KWh}}{\text{Kg T}}\right)=\frac{U.I.t}{1000.\ V.(C_i.TR)} \quad (5)$$

where C_i is the initial silica gel concentration (kg/m^3), I is the current intensity (A), U is the cell voltage (V), t is the treatment time (h), TR is the turbidity removal (%) and V is the volume of treated solution (m^3).

Optimisation by RSM

For optimisation of the EC–EF wastewater treatment assisted by OFI mucilage cladode. Four parameters were studied. The influence of each parameter was first separately investigated in single factor experiments (Table 1). To investigate the effect of OFI mucilage, the constant values for pH, voltage and conductivity were 7.67, 20 V and 1.31 mS/cm, respectively. Mucilage concentration was set at 2.5 mg/L, pH and conductivity were fixed at 7.67 and 1.31 mS/cm, respectively, to investigate the effect of voltage. To investigate the effect of pH: voltage, conductivity and mucilage concentration were set at 20 V, 1.31 mS/cm and 2.5 mg/L, respectively.

On the basis of single-factor experimental results, factor levels were chosen for each parameter. Then, an RSM based on a Box–Behnken design (BBD) (Manitab, trial version 10, USA) was conducted for optimisation (Table 2) (Adjeroud *et al*. 2015). Regression analysis of the data to fit a second-order polynomial equation (quadratic model) was carried out according to the following general equation:

$$Y=B_0+\sum_{i=1}^{k}B_iX_i+\sum_{i=1}^{k}B_{ii}X^2+\sum_{i>1}^{k}B_{ij}X_iX_j+E \quad (6)$$

Y represents the response function (in our case turbidity removal); B_0 is a constant coefficient; B_i, B_{ii} and B_{ij} are the coefficients of the linear, quadratic and interactive terms, respectively, and X_i and X_j represent the coded independent variables. According to the analysis of variance, the regression coefficients of individual linear, quadratic and interaction terms were determined. In order to visualise the relationship between the response and experimental levels of each factor and to deduce the optimum conditions, the regression coefficients were used to generate 3-D surface plots from the fitted polynomial equation. The factors levels were coded as -1 (low), 0 (central point or middle), $+1$ (high), respectively.

Analysis of variance was performed for the response variable using the full model where P-values (partitioned into linear and interaction factors) indicated whether the terms were significant or not. To verify the adequacy of the models, additional OFI mucilage-assisted EC–EF treatment trials were carried out at the optimal conditions predicted by the RSM and the obtained experimental data were compared to the values predicted by the regression model. Efficiency of EC–EF treatment aided by the OFI mucilage was evaluated based on the conditions selected by RSM on the turbidity removal.

Statistical analysis

Data obtained from the BBD were statistically analysed using ANOVA for the response variable in order to test the model significance and suitability, $P<0.05$ and $P<0.01$ were taken as significant and highly significant levels, respectively. BBD (Minitab, version 9.0.4.1, USA) software was used to construct the BBD model and to analyse all the results.

Table 1 Results of single-factor experiments for OFI mucilage assisted EC–EF process. Results are reported as means ± SD

Mucilage concentration (mg/L)	TR (%)	Voltage (V)	TR (%)	pH	TR (%)	Conductivity (mS/cm)	TR (%)
0	62.02 ± 11.50	15	80.61 ± 9.30	5	39.07 ± 1.42	1.80	90.36 ± 2.08
1	70.09 ± 12.20	20	89.97 ± 4.50	6	50.65 ± 6.83	2.00	87.78 ± 1.55
2.5	85.50 ± 02.00	25	81.18 ± 1.90	7	78.99 ± 0.30	2.50	93.93 ± 1.02
5	83.73 ± 03.26			8	80.68 ± 4.37	3.00	87.87 ± 3.68
7.5	78.12 ± 07.46			9	88.39 ± 2.24	3.40	85.98 ± 7.01
10	47.93 ± 00.55			10	64.11 ± 1.49		

Results and discussion

Single factors experiments

Influence of mucilage concentration on turbidity removal efficiency

Figure 2 shows the effect of mucilage concentration ranging from 1 to 10 mg/Lon turbidity removal. Maximum coagulation efficiency was observed at 2.5 mg/L with a turbidity removal of 85.5% (Fig. 2 and Table 1). With further increase in mucilage concentration, a significant reduction in the turbidity removal was noted.

The progressive evolution of turbidity removal with time as shown in Fig. 3 was kept constant and does not change after 20 min of the treatment for all tested mucilage concentrations.

In Fig. 3, at initial time, we note a negative turbidity removal which can be explained by the polymerisation of dissolved aluminium in the shape of aluminium hydroxide, thus increasing turbidity (Khemis *et al*. 2006). Fedala *et al*. (2015a) found that 0.2 mg/L of cactus concentration remove turbidity with 89% efficiency. The treatment of a synthetic wastewater prepared by Kaolin by Jar test and addition of 50 mg/L of *Moringa oleifera* seeds, 12 and 5 mg/L of mallow and okra mucilage reduce turbidity to reach 90, 93 and 96%

Table 2 Box–Behnken design (in coded and uncoded level of four variables) with the observed response and predicted values on the turbidity removal efficiency (TR%)

Run	Mucilage concentration (mg/L)	pH	Voltage (V)	Conductivity (mS/cm)	TR (%) Experimental	TR (%) Predicted
1	5 (+1)	7.00 (−1)	20 (0)	2.50 (0)	81.26 ± 4.82	80.94
2	5 (+1)	10.00 (+1)	20 (0)	2.50 (0)	89.50 ± 1.69	89.26
3	3 (0)	10.00 (+1)	20 (0)	3.00 (+1)	90.90 ± 1.49	90.88
4	1 (−1)	8.50 (0)	15 (−1)	2.50 (0)	85.05 ± 1.46	85.44
5	3 (0)	8.50 (0)	25 (+1)	3.00 (+1)	87.80 ± 1.15	87.47
6	3 (0)	8.50 (0)	20 (0)	2.50 (0)	91.00 ± 0.48	91.26
7	3 (0)	7.00 (−1)	15 (−1)	2.50 (0)	82.42 ± 1.49	82.38
8	1 (−1)	10.00 (+1)	20 (0)	2.50 (0)	90.41 ± 1.56	90.08
9	3 (0)	8.50 (0)	20 (0)	2.50 (0)	91.40 ± 0.01	91.26
10	1 (−1)	8.50 (0)	20 (0)	2.00 (0)	83.00 ± 2.47	83.86
11	3 (0)	8.50 (0)	15 (−1)	3.00 (+1)	88.06 ± 2.06	87.82
12	5 (+1)	8.50 (0)	20 (0)	3.00 (+1)	82.82 ± 3.40	83.10
13	3 (0)	8.50 (0)	25 (+1)	2.00 (−1)	88.66 ± 0.96	88.24
14	3 (0)	10.00 (+1)	20 (0)	2.00 (−1)	89.00 ± 0.67	88.59
15	1 (−1)	7.00 (−1)	20 (0)	2.50 (0)	84.57 ± 2.91	84.16
16	1 (−1)	8.50 (0)	20 (0)	3.00 (+1)	89.68 ± 0.23	89.56
17	3 (0)	10.00 (+1)	25 (+1)	2.50 (0)	91.00 ± 2.04	91.68
18	3 (0)	7.00 (−1)	20 (0)	2.00 (−1)	82.00 ± 0.70	82.00
19	5 (+1)	8.50 (0)	15 (−1)	2.50 (0)	85.88 ± 0.92	85.78
20	5 (+1)	8.50 (0)	25 (+1)	2.50 (0)	86.00 ± 0.80	85.59
21	3 (0)	8.50 (0)	20 (0)	2.50 (0)	91.38 ± 0.07	91.26
22	3 (0)	7.00 (−1)	20 (0)	3.00 (+1)	82.84 ± 0.49	83.23
23	3 (0)	10.00 (+1)	15 (−1)	2.50 (0)	90.00 ± 1.85	90.29
24	3 (0)	8.50 (0)	15 (−1)	2.00 (−1)	83.85 ± 5.06	83.52
25	5 (+1)	8.50 (0)	20 (0)	2.00 (−1)	85.00 ± 1.24	85.77
26	3 (0)	7.00 (−1)	25 (+1)	2.50 (0)	85.00 ± 7.45	85.35
27	1 (−1)	8.50 (0)	25 (+1)	2.50 (0)	89.90 ± 2.32	89.98

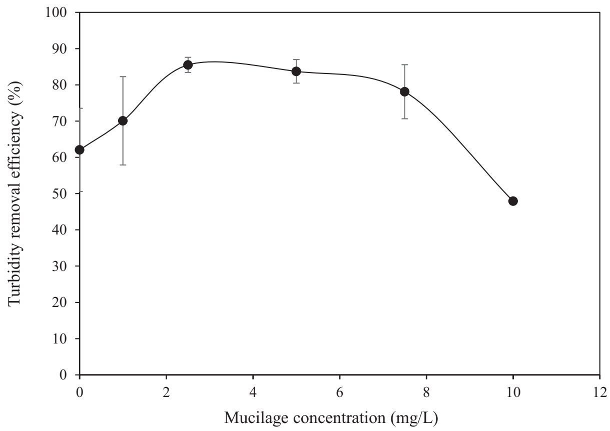

Fig. 2. Effect of initial concentration of OFI mucilage on turbidity removal efficiency: initial silica gel concentration $C_0 = 300$ mg/L, initial pH = 7.67, inter-electrode distance $d = 1$ cm, conductivity $k = 1.31$ mS/cm, treatment time $t = 20$ min, voltage $U = 20$ V.

efficiencies, respectively (Ndabigengesere *et al*. 1995; Anastasakis *et al*. 2009). Ramavandi *et al*. (2015) tested a proteinous coagulant extracted from Plantago ovata seeds on a turbid water and found that at lowest coagulant dosages (0.25–0.5 mg/L) the abatement of the turbidity was reduced to 99%.

Studies suggest that mucilage of the OFI, specifically its galacturonic acid component, may be the active component that provides coagulation activity by a mechanism which functions mainly through adsorption, separately galactose and rhamnose displayed no coagulation activity (Miller *et al*. 2008). In the okra mucilage, the presence of the hydroxyl groups (–OH) serves as active site for the attachment of the colloidal particles at mucilage (Freitas *et al*., 2015). Cactus mucilage has a similar composition, of the Okra mucilage; therefore, it is possible to consider that the mechanism is the same. FTIR result indicates that OFI mucilage contains a variety of functional groups such as carboxyl, hydroxyl, sulphate, phosphate, aldehydes, ketones and other charged groups (Fedala *et al*. 2015b). The functional groups present in the mucilage have been identified by FTIR spectra to be involved in pollutant adsorption (Nharingo & Moyo 2016).

In this study, it can be supposed that mucilage particles could interact directly with pollutants in solution, through bridging, prior to their adsorption to aluminium hydroxides. From another side, adsorption of the suspended molecules onto the surface of the aluminium species could facilitates their attachment to the OFI mucilage increasing then the coagulation efficiency as well as the turbidity removal. In both cases, small flocs form.

Influence of voltage on turbidity removal efficiency

Several studies suggest that the electrical voltage is one of the most important variable in EC–EF treatment. The high-current density increases bubble formation rate and decreases their size (Lakshmi & Sivashanmugam 2013).

Figure 4 shows the effect of voltage on turbidity removal efficiency at initial mucilage concentration of 2.5 mg/L. To investigate the effect of voltage on turbidity removal efficiency, different values of voltage were tested (15, 20 and 25 V).

According to Fig. 4, we note that turbidity removal reached 89.97% at 20 V, whereas at 15 and 25 V, the initial turbidity was diminished. This performance was similar to

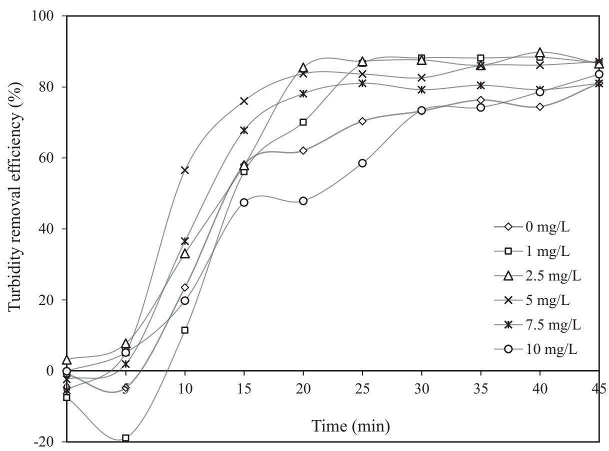

Fig. 3. Evolution of the effect of mucilage concentrations on turbidity removal efficiency during EC–EF for different values of initial mucilage concentrations: initial silica gel concentration $C_0 = 300$ mg/L, inter-electrode distance $d = 1$ cm, conductivity $k = 1.31$ mS/cm, voltage $U = 20$ V.

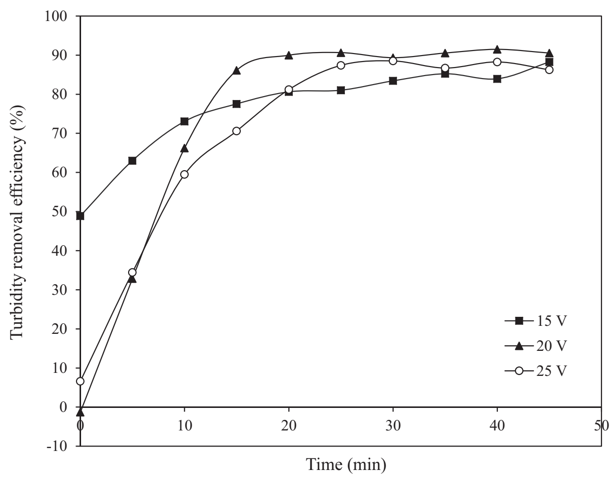

Fig. 4. Effect of applied voltage U on turbidity removal efficiency: initial mucilage concentration $C_m = 2.5$ mg/L, initial silica gel concentration $C_0 = 300$ mg/L, initial pH = 7.67, interelectrode distance $d = 1$ cm, treatment time $t = 20$ min, conductivity $k = 1.31$ mS/cm.

that found by Belkacem *et al*. (2008) where the turbidity removal found was 70.8% at 20 V.

The difference between the obtained results can be explained by the fact that at 15 V, formed bubbles had great size because the weak anodic dissolution of Al and H_2 gas production. Whereas, at 20 V, the anodic dissolution of Al increased, resulting in a greater amount of precipitate in parallel with a high production of H_2 bubbles gas then a better turbidity removal was observed. While, at 25 V, in spite of the high-current density, we note a low turbidity removal that can be interpreted by the noted increase of water temperature, which can be explained by the close distance between electrodes that affected the destabilisation of the ionic links formed between the pollutant and the coagulant.

Influence of pH on turbidity removal efficiency

In the electrochemical process, the pH is considered as an important parameter who must be controlled to lead to high efficacies (Lin & Chen 1997; Chen *et al*., 2000; Parsa *et al*., 2011; Secula *et al*. 2012).

Table 1 shows that at different pH levels ranging from 5 to 10, the EC–EF process is affected. The turbidity removal increases with pH; at alkaline medium, the turbidity removal was maximal at pH 9 with value of 88.39% (Table 1).

It was reported that in conventional EC process (EC–EF mucilage free treatment); the maximum turbidity removal was observed at pH around 8 (Merzouk *et al*. 2009), and in EC–EF treatment assisted by OFI pad juice turbidity removal was efficient at pH 8.7 (Adjeroud *et al*. 2015).

Studies report that the coagulation activity was dependent on the background electrolytes, at basic pH, the coagulation ability was increased at pH around 8 in the presence of electrolytes and at pH 10 without electrolytes (Miller *et al*. 2008).

According to Parsa *et al*. (2011), the pH affects $Al(OH)_3$ stability in the solution, in the high and low pH, $Al(OH)_3$ is in its charged form and is soluble in water, hence, it cannot be used for EC. But in neutral pH, $Al(OH)_3$ is stable and insoluble in the water and available for pollutant adsorption from water.

Llerena *et al*. (1996) found that the recovery of sphalerite was optimal at pH between 3 and 4 using buffer solution. They also documented that within this pH range, the hydrogen bubbles were the smallest, about 16 ± 2 μm. Typical bubble sizes in electrocoagulation always fall in the range of 20–70 μm they are far smaller than those observed in conventional air-assisted flotation (Adhoum *et al*. 2004).

Figure 5 shows the pH evolution according to time during the EC–EF treatment assisted by OFI mucilage. pH values tend towards neutrality whatever the initial pH of the solution. The ability to neutralise the pH of wastewater is a characteristic of EC–EF that was observed by several authors

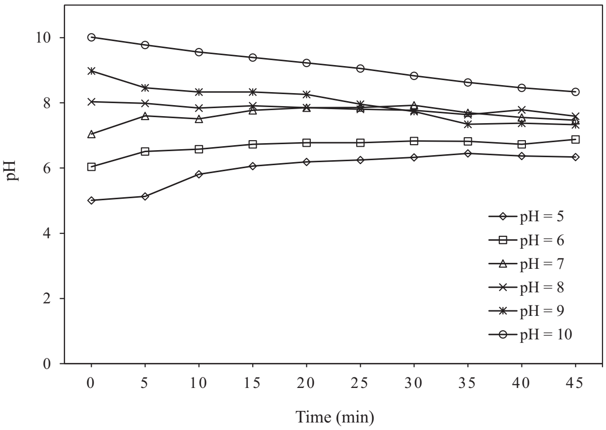

Fig. 5. Evolution of different initial pH values during EC–EF assisted with OFI mucilage: initial mucilage concentration C_m= 2.5 mg/L, initial silica gel concentration C_0= 300 mg/L, interelectrode distance $d = 1$ cm, conductivity $k = 1.31$ mS/cm, voltage $U = 20$ V.

(Chen *et al*. 2000; Kobya *et al*. 2006a; Merzouk *et al*. 2009; Adjeroud *et al*. 2015).

Thus, the pH value will be brought closer to neutral, where effective coagulation has been reported. This is explained by balance production between OH^- and their consumption, preventing high changes in pH (Chen *et al*. 2000; Chen 2004), which is advantageous for wastewater treatments.

Influence of conductivity on turbidity removal efficiency

Table 1 shows that different conductivity levels, ranging from 1.8 to 3.4 mS/cm, affect the EC–EF process. It is noted that elimination of turbidity reaches its maximum value of 93.93% at 2.5 mS/cm. The turbidity removal efficiency decreases to 87.78% and 85.98% at higher values of conductivity of 2.8 and 3.4 mS/cm, respectively.

This value is close to the value of 2.7 mS/cm the EC–EF reinforced by OFI pads juice for turbidity removal (Adjeroud *et al*. 2015).

In the EC–EF treatment, the conductivity affects the cell voltage and consumption of electrical energy. The increase of conductivity with electrolytes is known to reduce the Ohmic resistance of the wastewater to be treated and, therefore, the cell voltage at constant current density (Bayramoglu *et al*. 2004; Daneshvar *et al*. 2006; Kobya *et al*., 2006a). In addition, the energy consumption will decrease because it depends on cell voltage and current intensity.

Effect of OFI mucilage on SEEC

In all electrochemical processes, the electrical energy consumption is a significant parameter to the application of the

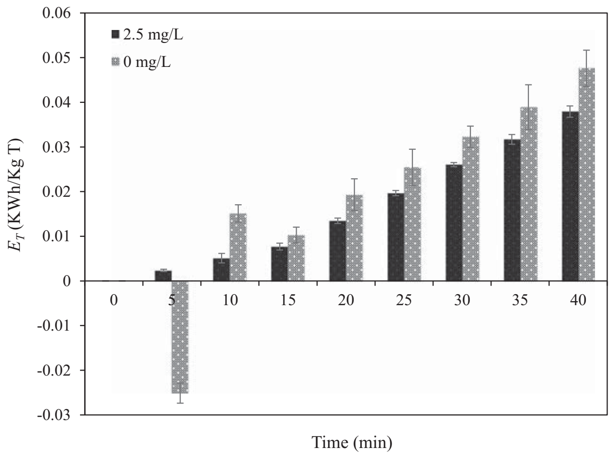

Fig. 6. Evolution of the specific electrical energy consumption with time in the absence (0 mg/L) and in the presence of OFI mucilage (C_m= 2.5 mg/L): initial silica gel concentration C_0= 300 mg/L, pH = 7.67, conductivity k = 1.31 mS/cm, voltage U = 20V.

process, mostly because it is related to the operating cost (Bayramoglu *et al*. 2004; Ghanbari *et al*. 2014).

The SEEC per kg turbidity removed (E_T) was calculated according to (Eq. (5)). Figure 6 displays the evolution of specific energy consumption with treatment time and mucilage. Energy consumption increases with time; however, it is slightly lower when mucilage concentration of 2.5 mg/L is added. This reduction is mainly shown at 40 min treatment time where the addition of 2.5 mg/L reduces the E_T from 0.4762 (EC–EF free mucilage treatment) to 0.3792 kWh/kg T. The negative value of E_T at 5 min is because of the negative value of TR as shown in Fig. 3.

Figure 7 shows the decrease of the specific energy consumption with the increase of turbidity removal, both related to the effect of mucilage. TR values represented on this figure are the same values of TR calculated using Eq. (4) and

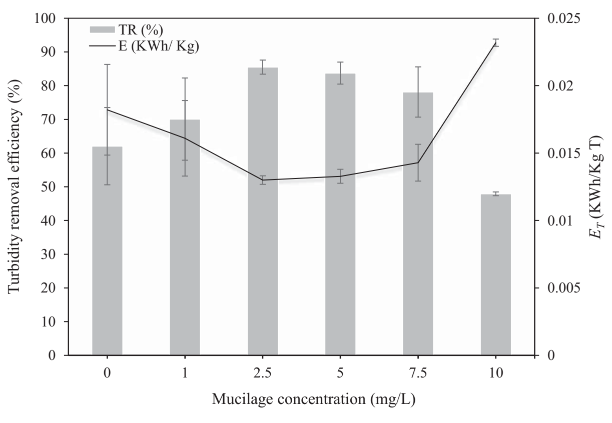

Fig. 7. Effect of OFI mucilage concentration on turbidity removal efficiency and on specific electrical energy consumption: initial silica gel concentration C_m= 300 mg/L, pH = 7.67, conductivity k = 1.31 mS/cm, voltage U = 20V, treatment time t = 20 min.

illustrated in Fig. 3. E_T values were calculated using Eq. (5) for all mucilage concentrations contrary to Fig. 6 where only 2.5 mg/L mucilage concentration was shown. Both values of TR and E_T correspond to treatment time of 20 min.

Apparently, OFI mucilage has the advantage to increase turbidity removal efficiency while decreases the specific energy consumption (Fig. 7) at a constant current density. For different heavy metals removal by EC–EF process without adjuvant addition the increase in current density led to a better EC removal efficiency but caused an increase in power consumption (Gatsios *et al*. 2015). Thus, OFI mucilage could withdraw the disadvantage of consuming more energy.

Optimisation by BBD

Modelling and fitting the model using surface response methodology

RSM is an effective widely used statistical technique for optimising complex processes, the main advantages of RSM are the reduced number of experimental trials needed to evaluate multiple parameters and their interactions (Ferreira *et al*. 2007). BBD is a class of three-level factorial designs. It has some advantages against the classical methods. It is a time- and money-saving method, because of lesser experiments. In addition, this method is able to show the main and interaction effects of variables (Ashrafi *et al*. 2016).

Following the results of single factor experiments (preliminary trials reported in Table 1), the chosen parameter ranges were equal to 1–5 mg/L for mucilage concentration, 7–10 for pH, 15–25 V for voltage and 2–3 mS/cm for conductivity. The design matrix and the corresponding results of RSM experiments to determine the effects of the four independent variables including mucilage concentrations (X_1), pH (X_2), voltage (X_3) and the conductivity (X_4) on the turbidity removal efficiency obtained by EC–EF treatment assisted by OFI mucilage in the trials of the BBD are reported in Table 2. While Table 3 reports the statistical analysis of the regression model (Eq. (7)).

Including significant and non-significant terms, the following predictive mathematical equation was obtained:

$$\begin{aligned} Y_{(TR)} = {} & 91.26 - 1.01X_1 + 3.56X_2 + 1.09X_3 + 0.88X_4 + 0.60X_1X_2 \\ & - 1.18X_1X_3 - 2.22X_1X_4 - 0.39X_2X_3 + 0.27X_2X_4 \\ & - 1.27X_3X_4 - 2.94X_1^2 - 2.21X_2^2 - 1.62X_3^2 - 2.87X_4^2 \end{aligned} \quad (7)$$

It can be seen that all the linear parameters (X_1, X_2, X_3, X_4) and their quadratic parameters were highly significant at the level of $P < 0.01$. The interaction parameter (X_1X_2)was significant ($P < 0.05$) and highly significant in the case of (X_1X_3),

Table 3 Estimated regression coefficient for the quadratic polynomial model and the analysis of variance (ANOVA) for the experimental results

Parameter[a]	Estimated coefficients	Standard error	DF[b]	Sum of squares	*F*-value	Prob >*F*
Model intercept	91.26	0.30	14	293.18	80.2	0.0001
B_0	91.26	0.30	1	293.18	80.2	<0.0001
Linear						
X_1	−1.01	0.15	1	12.30	47.11	<0.0001
X_2	+3.56	0.15	1	152.08	582.4	<0.0001
X_3	+1.09	0.15	1	14.30	54.76	<0.0001
X_4	+0.88	0.15	1	9.35	35.79	<0.0001
Quadratic						
X_1^2	−2.94	0.22	1	46.03	176.29	<0.0001
X_2^2	−2.21	0.22	1	26.03	99.68	<0.0001
X_3^2	−1.62	0.22	1	13.98	53.55	<0.0001
X_4^2	−2.87	0.22	1	43.94	168.28	<0.0001
Interaction						
X_1X_2	+0.6	0.26	1	1.44	5.51	0.03680
X_1X_3	−1.18	0.26	1	5.59	21.42	0.00060
X_1X_4	−2.22	0.26	1	19.62	75.15	<0.0001
X_2X_3	−0.39	0.26	1	0.62	2.39	0.14810
X_2X_4	+0.27	0.26	1	0.28	1.08	0.32010
X_3X_4	−1.27	0.26	1	6.43	24.61	0.00030
Lack of fit			10	3.03	5.97	0.15190
Pure error			2	0.10		
Residual			12	3.13		
R^2					0.9894	
R^2_{adjusted}					0.9771	
C.V. %	0.59					
RMSE	0.5110					
Cor total			26	296.32		

[a]Coefficient refer to the general model.
[b]Degree of freedom.
X_1: OFI mucilage, X_2: pH, X_3: voltage, X_4: conductivity.

(X_1X_4) and (X_3X_4) at the level of $P < 0.01$, whereas (X_2X_3) and (X_2X_4) were non-significant ($P > 0.05$) (Dahmoune *et al*. 2013; Adjeroud *et al*. 2015).

Eq. (7) is used to obtain the response surface for all the possible variables interactions (Fig. 8). The turbidity removal efficiency reached a maximal level at the medium value of mucilage concentration, voltage, pH and conductivity (0 coded values) (Table 2).

Table 3 reports the analysis of variance (ANOVA) for the experimental, the model fitted *F-value* of 80.20 indicates that the model is significant and there is only 0.01% chance that a "Model *F-value*" this large could occur because of noise. The determination coefficient R^2 was 0.98, which implies that sample variations of 98.9% for the OFI mucilage assisted EC-EF treatment efficiency were attributed to the independent variables, and only 1.1% of the total variations cannot be explained by the model. In a good statistical model, $\mathbf{R^2_{adjusted}}$ should be comparable and close to R^2 (Dahmoune *et al*. 2015), that what we observed in Table 3.

A confirmation of the validity of the model is the Lack of Fit. A lack of fit "*F-value*" of 5.97 implies that the Lack of it is not significant relative to the pure error ($P > 0.05$). There is 15.19% chance that Lack of Fit "*F-value*" this large could occur because of noise.

The coefficient of variation (C.V.%) was equal to 0.59% and the "Adequate Precision" ratio, not shown in the table, of 28.218 suggests that the model is reliable and reproducible agreeing previous reports (Chen *et al*. 2012). In general, a C.V. higher than 10% indicates that variation in the mean value is high and does not satisfactorily develop an adequate response model (adequate signal for the model) (Karazhiyan *et al*. 2011). The results showed a suitable signal for the model suggesting that it could work well for water treatment by EC–EF technique assisted by OFI mucilage.

Analysis of RSM

The effects of the independent variables and their mutual interactions on the turbidity removal efficiency can be visualised through the three-dimensional (3D) response surface plots shown in Fig. 8. Each 3D plot represents the number of combinations of the two-test variable. 3D response surface is the graphical representations of regression equation and is very useful to evaluate the relationship between independent and dependent variables.

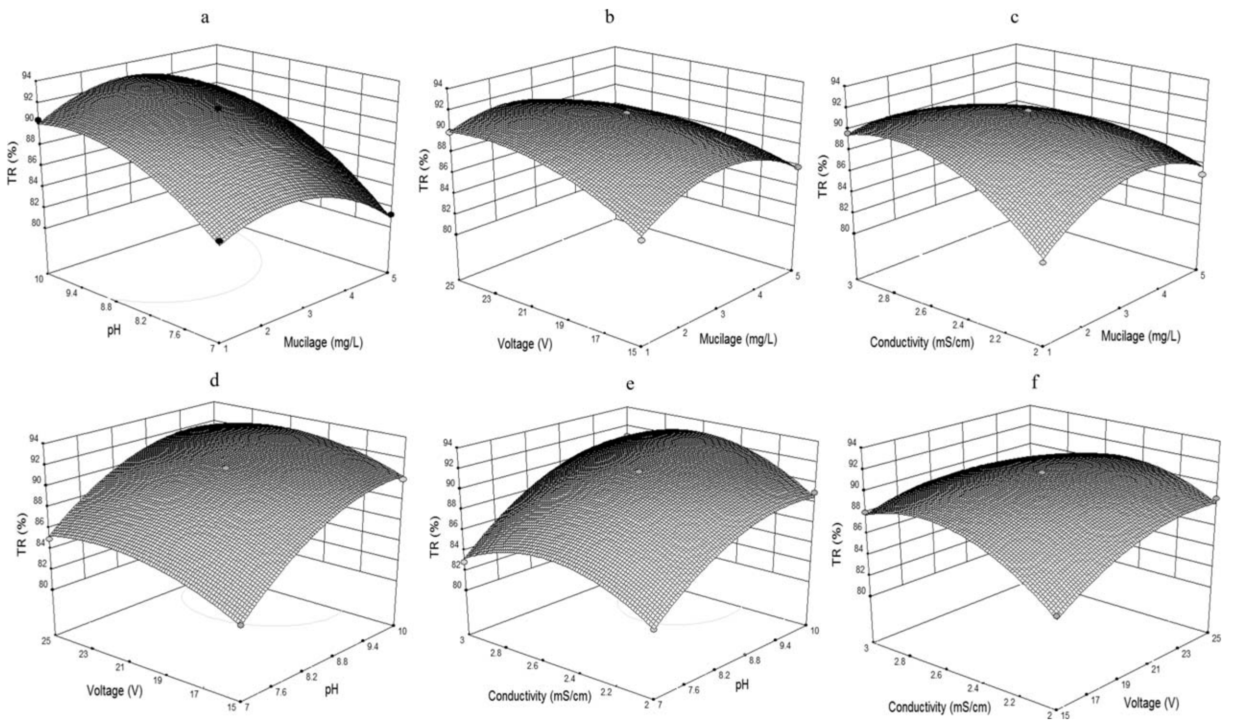

Fig. 8. Response surface plots (a–f) showing the interactive effects of (a) pH and mucilage concentration, (b) voltage and mucilage concentration, (c) conductivity and mucilage concentration, (d) voltage and pH, (e) conductivity and pH, and (f) conductivity and voltage on the turbidity removal efficiency. Experimental data and conditions are shown in Table 2.

Figure 8(a–c) depicts the interaction between OFI mucilage concentration and each of the three other factors (voltage, pH and conductivity) on the EC–EF treatment. They show that turbidity removal efficiency increases with increase of the voltage and conductivity at their medium values, it rises too with the increase in pH and decrease of OFI mucilage concentration. In Fig. 8(d and e), the turbidity removal efficiency increased gradually to reach its maximal value with increasing pH and voltage values, and pH and conductivity values, respectively. While in Fig. 8(f), the recovery of turbidity was increased at the medium values of conductivity and voltage.

As shown in Table 3, the turbidity removal efficiency principally depends on the OFI mucilage concentration, pH, voltage and conductivity as their linear and quadratic effects were highly significant ($P < 0.01$). However, the interaction effects of OFI mucilage concentration with voltage and conductivity were highly significant ($P < 0.01$), as well as the interactive effect of voltage and conductivity.

Validation and verification of the predictive model

The results experiments performed to optimise turbidity removal efficiency using EC–EF had the following critical values: OFI mucilage concentration 2.5 mg/L, pH 9.65, conductivity 2.61 mS/cm and voltage 21.2 V.

The appropriateness of the model equation for predicting the optimum response values was tested using the selected optimal conditions. The predicted turbidity removal efficiency value was equal to 92.96% ± 0.01, which, was consistent with the practical turbidity removal value of 93.14 ± 1.31. Also, the predicted values were in close agreement with the experimental values (Table 2) and were found to be not significantly different at $P > 0.05$ using a paired *t*-test (Hossain *et al*. 2012).

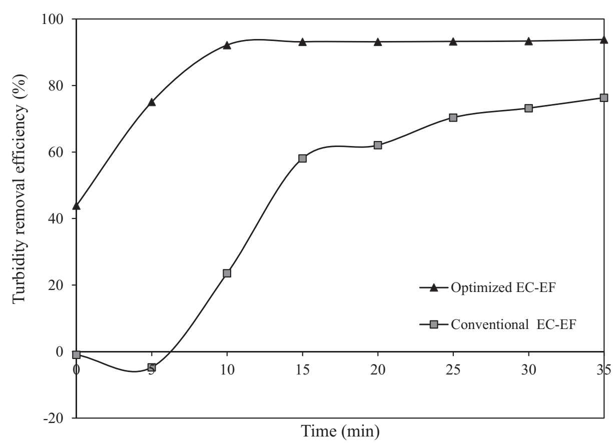

Fig. 9. Effect of RSM optimisation on turbidity removal efficiency. Conventional EC-EF: pH = 7.67, conductivity $k = 1.31$ mS/cm, voltage $U = 20$ V. Optimized EC–EF OFI mucilage $C_m = 2.5$ mg/L, pH = 9.65, conductivity $k = 2.61$ mS/cm, voltage $U = 21.2$ V.

Figure 9 shows the comparison of turbidity removal efficiencies obtained by RSM optimised EC–EF assisted by OFI mucilage and conventional EC–EF (EC–EF free mucilage treatment). Turbidity removal efficiency during conventional EC–EF was equal to 62.02% at 20 min treatment time and that of OFI mucilage assisted EC–EF equal to 93.14% ± 1.31 for the same time. The RSM optimisation in the presence of OFI mucilage allows an enhancement of the electrochemical process by 30.94%.

Conclusion

(1) A biodegradable natural coagulant extract from OFI cladodes; mucilage, was introduced to the EC–EF treatment technique, in order to ameliorate its efficiency. The orientation towards natural substances helps restraining the toxicity of chemicals, potentially added to EC–EF, on human health and on the environment as well as reducing the cost of industrial effluent treatment.

(2) The RSM was successfully employed to optimise the EC–EF process. Under the optimal operating conditions, the turbidity removal was enhanced by 30.94%. Furthermore, the RSM was useful to investigate the effect of the four studied parameters (OFI mucilage concentration, pH, conductivity and voltage) on the turbidity removal response. The applied second-order polynomial model gave a satisfactory description of the experimental data; it showed that turbidity removal efficiency was affected by the forth-studied parameter. It is important to note the reduction in EC–EF specific electrical energy consumption because of the presence of the bio-coagulant, this could be very promising for small or large-scale applications.

Conflict of interest

The authors declare that there are no conflicts of interest.

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