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Removal of color caused by dissolved organic matter from groundwater by electroflotation-filtration continuous flow reactor and optimization by response surface methodology

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ABSTRACT

To decrease the color mainly caused by the dissolved organic matter (DOM) in underground water with low turbidity and high chromaticity, a coagulation and electroflotation-filtration process was proposed. The investigated operating parameters based on minimum residual color were coagulant type, dose, electrodes materials, filtering velocity, and current intensity. The electroflotation–filtration reactor was operated in the vertical continuous flow mode. The best coagulant was polymerization ferric chloride (PFC). Aluminum electrodes were selected as anodes by comparison experiments. The results indicated that the increase in filtering velocity resulted in decrease in color removal and shorter standard processing time with the aluminum electrode. When the current was below a certain value, increasing current intensity shortened the time to reach the minimum residual color and achieved higher color removal. The interaction between the factors and their optimum levels for minimum residual was determined using response surface methodology. The optimum operation conditions were the filtering velocity of 5.00 m/h, the PFC dosage of 36.07 mg/L, and current intensity of 2.00 A with aluminum electrode, and the minimum residual color of 4.52 Pt–Co units could be obtained.

Keywords: Continuous flow reactor; Dissolved organic matter; Electroflotation; Groundwater; Response surface methodology

1. Introduction

Groundwater provides huge drinking water sources, and most of the time, avoided by elevated levels of color. The color of groundwater can be caused by dissolved organic matter (DOM) and some metal ions, such as Fe^{2+} and Mn^{2+}. The presence of color is loss of public confidence in the quality of potable water and the World Health Organization (WHO) guidelines give a guide level of 15 Pt–Co units for color [1].

Although many literatures reported on removal Fe^{2+} and Mn^{2+} [2–4], few assays devoted to remove color caused by DOM for decolorization of groundwater. Coagulation is a traditional water treatment method. And hydrophilic and lower molecular weight

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DOM is less effectively removed by coagulation than hydrophobic and higher molecular weight DOM [5]. It is similar to sedimentation and filtration [6]. Typical processes applied to remove DOM of groundwater include ozone oxidation [7], activated carbon adsorption [8], and nanofiltration [9]. Ozone oxidation forms by-products which bring out health risk. High molecular organic compounds, such as humic and fulvic acids, which may cause color in groundwater [10], are known to be important precursors for disinfection by-products [11]. While both nanofiltration and activated carbon adsorption are costly. The most widely used methods, such as traditional coagulation, flotation, adsorption, and chemical oxidation or combination of these are insufficient somewhile for groundwater of low turbidity and high chromaticity treatment. Therefore, new process should be exploited to treat groundwater for organic color removal.

Separation by flotation is relatively simple and faster compared to coagulation and sedimentation. Janssen and Koene [12] had shown that electrochemical techniques were one of the competitive and interesting technologies in this field. Electroflotation techniques are highly versatile and are more competitive than other flotation techniques such as dissolved air flotation and dispersed air flotation for treatment of pollutants [13,14]. Weintraub et al. [15] considered an electrolytic process to treat oily wastewater. In Mansour's research, the coagulation-electroflotation process was used to treat paper industry effluents which contain an important rate of suspended solids [16]. And the coagulation-electroflotation process was also used to treat cardboard industry effluents having a high COD, BOD, and suspended solids charge [17]. Moreover, Mohora E et al. [18] used electrocoagulation/flotation to remove natural organic matter and arsenic from water. Other literatures also reported the removal of dissolved organic carbon (DOC) by electrocoagulation [19,20].

Electroflotation is the flotation using electrolytically generated bubbles of hydrogen and oxygen for separating suspended substances from aqueous phases, and widely used in wastewater treatment [21]. It has been applied to the removal of dyes [22,23], restaurant wastewater, [24] turbidity, and heavy metals [25]. Compared with the conventional dissolved air flotation, electroflotation has many advantages, including high flotation efficiency, compact units, easy operation, and less maintenance.

This study based on actual case study in a waterworks in North China. The traditional process of coagulation-sedimentation-filtration of the plant cannot reach the effluent standard in color. Process of coagulation followed by electroflotation-filtration was exploited to treat groundwater for organic color removal. Compared with traditional treatment, this study combined coagulation, electrocoagulation/flotation, and filtration. Furthermore, the reactor was conducted in continuous mode. Effect of electrode material, filtering velocity, current intensity, and coagulation was studied and optimized by response surface methodology (RSM). All of this would be helpful for the treatment of groundwater.

2. Materials and method

2.1. Raw water characteristics

The raw water used in the study was obtained from underground water-bearing deposits. The influent characteristics (averages) during the experiments are presented in Table 1 provided by Wuqinglongquan Water Plant. It indicates that the heavy metal is not the material cause of color in groundwater because of the low concentration of Fe^{2+} and Mn^{2+}.

2.2. Microfiltration-ultrafiltration-ozonation test

The pore size of membrane for microfiltration (PVDF, Pall Co., USA) and ultrafiltration (Dizzer 5,000, Inge Co., Germany) was 0.1 μm and 0.015 μm, respectively. The ultrafiltration filter has a nominal molecular weight cut-off of 100 kDa. The raw water was disposed by microfiltration (trans-membrane pressure 0.7 bar), ultrafiltration (trans-membrane pressure 0.8 bar), and ozonation sequentially ($10 mgO_3/L$, 10 min).

2.3. Jar test of coagulation

Coagulation experiments were operated in a coagulation-flocculation test mixer (JJ-6). The applied coagulants are polymeric ferric sulfate (PFS) (Fe content ≥18.5%, basicity = 0.09–0.14), polymerization ferric chloride (PFC) ($FeCl_3$ content ≥ 30%), polyaluminium

Table 1
Characteristics of the raw groundwater

Parameters	Values
Turbidity (NTU)	1.2 ± 0.1
Color (Pt–Co units)	38 ± 1
TOC (mg/L)	2.91 ± 0.12
UV_{254} (cm^{-1})	0.0484 ± 0.0037
COD (mg/L)	2.65 ± 0.22
pH	7.2 ± 0.2
DO (mg/L)	6.32 ± 0.11
Fe (mg/L)	0.08 ± 0.002
Mn (mg/L)	0.006 ± 0.001

chloride (PAC) (Al_2O_3 content ≥27%, basicity = 0.45–0.96), and polyaluminium ferrous chloride (PAFC) (Al_2O_3 content ≥29%, Fe_2O_3 content = 3–5%, basicity = 0.65–0.92). And the doses of each coagulant ranged from 10 to 70 mg/L. The time of coagulation and sedimentation was determined according to the running time of plant. In all of the coagulation experiments, 300 mL of groundwater was rapidly mixed (100 rpm) for 1 min, then mixed at 80 rpm for 5 min, and finally slowly mixed (30 rpm) for 10 min with various dosages of different coagulants. The water was allowed to settle for 15 min and the supernatant was taken to be filtered for measuring color.

2.4. Electroflotation-filtration experiments

The water used for continuous filtration was from flocculating chamber of the water plant. Three reactors were adopted for downward filtration as shown in Fig. 1, which are 4.0 m high plexiglass column, with 18.5 cm inner diameter. The bottom was 1.2-m high quartz sand filter (0.55–0.92 mm diameter, 2,650 kg/m^3 density, coefficient of graining non-uniformity K80 = d80/d10 < 2.0). As for electroflotation, metal electrode rings were installed at the inlet of the column with continuous direct current. Influent and effluent quality of the water was tested.

2.5. Analysis methods

Water pH was measured by an 828 Orion pH meter. Turbidity was analyzed with a 2100P Hach photoelectric turbidity meter, and the color was determined by Hach LICO 500 Colorimeter.

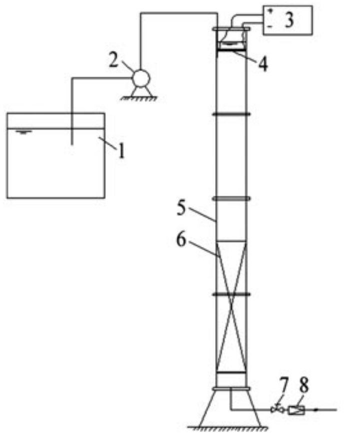

Fig. 1 Schematic of electroflotation filtration unit in continuous mode: (1) sedimentation tank, (2) upgraded pump, (3) direct current power, (4) electrolytic coil, (5) filtering cylinder, (6) silica sand filtering medium, (7) effluent regulating valve, (8) effluent flow meter.

2.6. Optimization by response surface methodology

The statistical analysis that involves the determination of the optimum levels of significant variables for the removal of DOM was performed using RSM with Box–Behnken design (BBD). The variables involved PFC dosage, filtering velocity, and current intensity. Each of the variables was assessed at three coded levels, −1, 0, and +1 (Table 2). A total of 17 experiment runs were conducted (Table 3). The full experimental plan with regard to their values in coded form is provided. All the experimental design and statistical analysis of the data were done by using Design-Expert version 8.0.6.

3. Results and discussion

3.1. Microfiltration-ultrafiltration-ozonation test

It can be concluded that Fe^{2+} and Mn^{2+} are not the reason of high color, since the concentration of Fe^{2+} and Mn^{2+} is rather low. By microfiltration-ultrafiltration-ozonation test, the removal rates of the color in raw groundwater were 23, 34, and 23%, respectively. It indicates that complicated factors contribute to high color of the groundwater and organic matter is one of the dominant factors. The increase in DOM makes the colloid steady. Increasing coagulant dosage guarantees rather short normal cycle with frequent backwash and cannot improve effluent quality obviously in the plant.

3.2. Determination of type and dose of coagulant for optimized coagulation

Coagulation is the most important stage in drinking water treatment processes for the maintenance of acceptable treated water quality. In practice, the required coagulant dosage in water treatment is usually evaluated by jar tests [26]. The comparison of

Table 2
The factors and levels of response surface methodology experiments

			Code level values		
Factor	Name	Units	−1	0	1
A	PFC dosage	mg/L	30	40	50
B	Filtering velocity	m/h	5.0	5.5	6.0
C	Current intensity	A	0.50	1.25	2.00

Table 3
The results of response surface methodology experiments for Box–Behnken design

No.	*A*	*B*	*C*	Residual color
1	0	−1	1	5
2	0	1	1	9
3	1	0	−1	15
4	0	0	0	9
5	−1	0	1	11
6	0	0	0	11
7	0	0	0	11
8	0	0	0	11
9	0	0	0	11
10	1	0	1	17
11	−1	−1	0	11
12	−1	0	−1	18
13	1	1	0	17
14	1	−1	0	15
15	0	−1	−1	8
16	0	1	−1	14
17	−1	1	0	19

commercially available polymeric ferric sulfate (PFS), polymerization ferric chloride (PFC), polyaluminum chloride (PAC), and polyaluminum ferrous chloride (PAFC) was conducted at pH 7.

The effect of various coagulant concentrations of four coagulants on the residual color of the raw water is shown in Fig. 2. The results revealed that the tests achieved the lowest residual color at the dosages of 40 mg/L PFS, 40 mg/L PFC, 15 mg/L PAC, 25 mg/L PAFC, respectively. Thereinto, the most optimal color removal efficiency during the jar tests could achieve at 40 mg/L dosage of PFC. It can be seen from Fig. 2 that residual color was much lower using PFC as coagulant than using others. In addition to this, the needed dosage of PFC was similar with other coagulants. Considering the removal efficiency and the cost, PFC was chosen as the best coagulant for the lowest residual color achieving. Additionally, other studies showed that PFC performed excellent efficiency for pollutants removal using enhanced coagulation [27,28]. One of them reported that the color removal efficiencies were unsatisfactory when the dosage of PFC varied from 10 to 20 mg/L, due to the reason that zeta potential was negative (zeta potential varied from −10.03 to −3.17 mV at PFC dosage varied from 10 to 20 mg/L) under lower dosage of PFC so that coagulants could not have the ability to attract pollutants, and also the flocs might not form well [29]. Increasing PFC dosage caused increase in color removal efficiency because the formed flocs structures were more enhanced and compact [29]. However, the removal efficiency of color decreased when PFC dosage further increased higher than 40 mg/L, which was attributed that overmuch PFC dosage could lead the flocs to lose structures because of smaller flocs dimension under higher dosage.

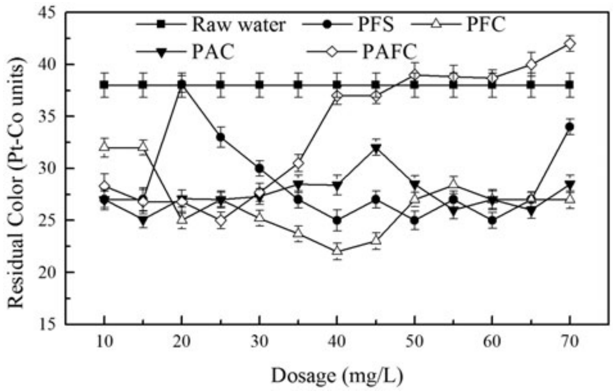

Fig. 2. Effect of coagulant dose and type on residual color for groundwater.

The residual turbidity of water was shown in Table 4, revealing that all the four coagulants could make the turbidity under 0.8 NTU, which could satisfy the standard. The coagulants removed the turbidity by charge neutralization and absorption-bridging ability. The low level of turbidity of raw groundwater may be the reason that effects of different coagulants were essentially the same. In addition, the pH of water decreased slightly due to the weak hydrolysis of PFC.

3.3. The comparison of anode materials

Electro-coagulation-flotation (ECF) has been reported as an alternative to water treatment processes. The electrolysis of water generates electrolytic oxidation (or reduction), electrolytic coagulation, and electrolytic flotation, using some metals as anode material. So electroflotation process is also ECF process in this study. Electrode material plays an important role in ECF process. However, few researches pay attention to the comparison of different electrode materials on the color removal of groundwater. Aluminum and iron are widely used electrode materials in ECF process [30–32]. In this study, comparison of aluminum and iron used as anode material was taken into consideration, under the same operation conditions. The treatment effect of the aluminum electrode was better than the iron electrode, as it was shown in Fig. 3. The time

Table 4
The residual turbidity of water under different kinds and dosages of coagulants

	PFS		PFC		PAC		PAFC	
	Turbidity (NTU)		Turbidity (NTU)		Turbidity (NTU)		Turbidity (NTU)	
Dosage(mg/L)	Raw water	Effluent	Raw water	Effluent	Raw water	Effluent	Raw water	Effluent
10	1.23	0.42	1.30	0.65	1.28	0.57	1.15	0.23
15	1.20	0.21	1.23	0.43	1.21	0.62	1.17	0.22
20	1.17	0.50	1.18	0.88	1.15	0.33	1.23	0.57
25	1.15	0.22	1.19	0.38	1.22	0.71	1.18	0.49
30	1.23	0.35	1.21	0.21	1.24	0.40	1.21	0.21
35	1.22	0.14	1.21	0.19	1.16	0.53	1.20	0.55
40	1.24	0.09	1.22	0.10	1.12	0.21	1.14	0.28
45	1.20	0.15	1.19	0.17	1.18	0.46	1.17	0.25
50	1.19	0.25	1.16	0.96	1.26	0.43	1.22	0.54
55	1.22	0.13	1.17	0.86	1.23	0.66	1.25	0.21
60	1.21	0.08	1.18	0.37	1.15	0.57	1.21	0.37
65	1.22	0.15	1.21	0.45	1.23	0.47	1.23	0.41
70	1.18	0.09	1.20	0.27	1.17	0.44	1.16	0.53

of the effluent color below 15 Pt–Co units was 16 h for aluminum electrode, which was only 13 h for iron electrode. And aluminum electrode tended to be more efficient than iron electrode by achieving a higher removal rate of color. The standard processing time is longer, and in the majority of color removal period with aluminum electrodes than iron electrodes for many reasons.

The main electrolysis reactions are as follows in the ECF process using aluminum and iron electrodes:

When aluminum electrode is used,

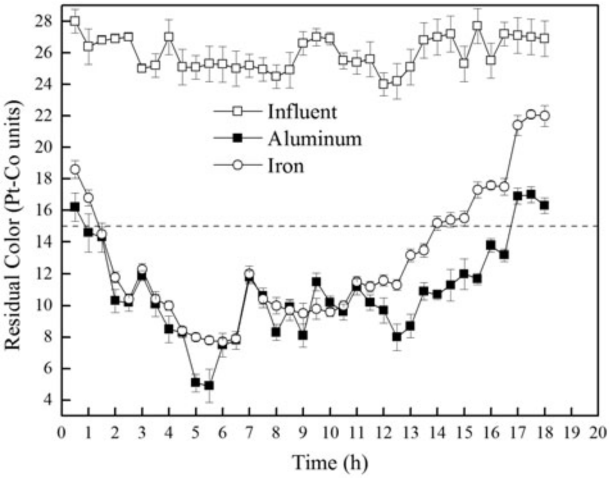

Fig. 3. Residual color as a function of treatment time using different electrode materials. Condition: current intensity, 1 A; initial pH 7; filtering velocity, 5.0 m/h.

The release of Al^{3+} ions at the anode accompanied by the oxidation of water:

$$Al \rightarrow Al^{3+} + 3e^- \tag{1}$$

$$2H_2O \rightarrow 4H^+ + O_2 + 4e^- \tag{2}$$

In solution:

$$Al^{3+} + 3H_2O \leftrightarrow Al(OH)_3 + 3H^+ \tag{3}$$

The reduction of water at the cathode:

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \tag{4}$$

When iron electrode is used,

The release of Fe^{2+} ions at the anode accompanied by the oxidation of water:

$$Fe \rightarrow Fe^{2+} + 2e^- \tag{5}$$

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \tag{6}$$

In solution:

$$Fe^{2+} + 2OH^- \leftrightarrow Fe(OH)_2 \tag{7}$$

$$4Fe^{2+} + 4H^+ + O_2 \rightarrow 4Fe^{3+} + 2H_2O \tag{8}$$

$$Fe^{3+} + 3OH^- \leftrightarrow Fe(OH)_3 \tag{9}$$

The reduction of water at the cathode:

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \qquad (10)$$

When aluminum and iron are used as anode, respectively, the anodic electrolysis products are mainly Al^{3+} and Fe^{2+} (Eqs. (1), (2), (5), and (6)), which further immediately hydrolyze to produce hydroxides and polyhydroxides with H_2O, OH^-, and O_2 in the water (by the Eqs. (3), (7)–(9)) with the release of H_2 at the cathode. The hydroxides and poly hydroxides were reported to have stronger capacity to capture the pollutants that caused color in water, which resulted in more coagulation in the water. Moreover, the existence of Fe^{3+} might inhibit the electrolysis of anode with iron, which led to the decrease in color removal in groundwater. On the other hand, the Fe hydroxides and poly hydroxides have denser texture than Al hydroxides and polyhydroxides, attaching to the electrode after the formation, then reducing the electrolysis efficiency. Thereby decrease in the yield of H_2 occurred and removing chromaticity capacity weakened. Also, higher current efficiency was generated in the case of aluminum electrodes than iron electrodes, which resulted in the higher removal of color with aluminum electrodes [33]. The similar results were revealed by Gao et al. [34], which reported that iron electrodes were observed to be less efficient as compared with aluminum electrodes, as demonstrated by the difference between the removal efficiencies (78.9% vs. 100%) at 45 min. In addition, another reason leading to the better removal of color with aluminum electrodes was that the amount of Al hydroxides and polyhydroxides was much higher than that of Fe hydroxides and polyhydroxides under the pH of 7 [35]. Overall, aluminum was promising for the color removal in groundwater and selected as the anode material in the following experiments.

3.4. *The effect of filtering velocity (v)*

The impact of filtering velocity on the color removal efficiency of groundwater on ECF-filtration reactor was investigated by carrying out experiments with aluminum electrode at filtering velocity values: 5.0 m/h, 5.5 m/h, and 6.0 m/h, while the other parameters were kept constant ($I = 1A$, initial groundwater pH 7.0).

The influence of filtering velocity on effluent pH was also studied. Al^{3+} ions were released into water during the process. Some Al^{3+} was adsorbed by organic matters for neutralizing the negative charge. On the other hand, hydrolysis of Al^{3+} consuming Al^{3+} ions. In general, increase in effluent pH slowed as filtering velocity increased.

Fig. 4 shows the effect of various filtering velocities on the residual color of groundwater. The filtering velocities show a strong effect on the residual color of groundwater and the minimum residual color reached 5.1 Pt–Co units, 5.2 Pt–Co units, and 7.2 Pt–Co units, respectively, depending on the filtering velocity values (5.0–6.0 m/h). Similarly, the filtering velocities have a large effect on the time of duration of the residual color reaching the standard. The reactor with lower filtering velocity exhibited advantages. For the filtering velocities of 5.0 m/h, 5.5 m/h, and 6.0 m/h, the duration time that the residual color below 15 Pt–Co units were 16 h, 13 h, and 8 h, respectively. The reason of this phenomenon might be the water residence time was longer when the filtering velocity was smaller, which resulted in the existence of more microbubbles in the same volume water. Hence, more flocculated particles were removed by lifting to the surface with the microbubbles adhered to them. Meanwhile, the groundwater had sufficient time to be treated. For the purpose of removing color from underground water, only operation parameters of the treatment have been investigated. Furthermore, the quality of water is relatively stable, thus, the effects of other parameters of water quality changing were not carried out. In addition, the efficiency of filtration was higher with lower filtering velocity. However, Mohora E observed that the flow rate does not appear to have a large effect on natural organic matters removal by ECF reactor and

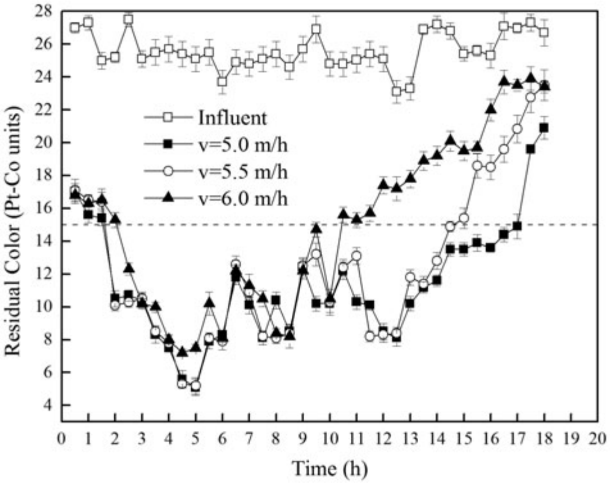

Fig. 4. Residual color as a function of treatment time with different filtering velocities in electroflotation-filtration reactors using aluminum electrodes. Condition: current intensity, 1 A; initial pH 7.

after 15–90 min of treatment and the average UV_{254} absorbance reductions were 72–77% [18].

3.5. The effect of current intensity (I)

It is considered that current density plays a very important role in any electroflotation process as it has a significant influence on reaction kinetics and determines the pollutant removal efficiency, specific energy, and energy consumption [25,36]. In this study, the aluminum coil was used as anode, which made it difficult to measure the superficial area. Therefore, current intensity was investigated instead of current density. The experiments were conducted at current intensity values: 0.5 A, 1.0 A, 1.5 A, 2.0 A while filtering velocity, number of electrodes, and initial groundwater pH were kept constant ($v = 5.0$ m/h, initial pH 7) to study the color removal and optimize the current intensity.

Residual color as a function of electrolysis time at different current intensities was shown in Fig. 5. At the initial stage, the residual color decreased with the increase in current intensity. It appears that, despite the different residual color, there is a resemblance among curves when the current intensity was 0.5 A and 1.0 A. The residual color decreased gradually with the operating time and reached relative equilibrium within 35–45 min. Similarly, the curves had a similar trend when the current intensity was 1.5 A and 2.0 A. A rapid reduction in residual color of groundwater was followed by an andante decrease and achieved steady state. The residual color reached 8 and 7 Pt–Co units with the current intensity of 0.5 A and 1.0 A, while it decreased to 6 Pt–Co units when the current intensities were 1.0 A, 1.5 A, 2.0 A, respectively. Effect of current intensity on pH was also analyzed. The increase in current intensity from 0.5 A to 2.0 A resulted in a greater increase in effluent pH. The higher current intensity released more Al^{3+} ions made a faster increase in effluent pH.

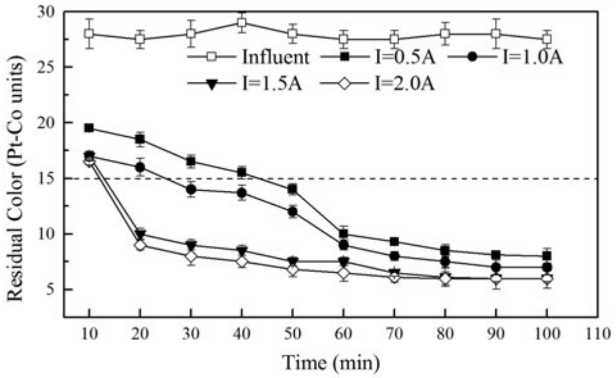

Fig. 5. Residual color as a function of treatment time at different current intensity in electroflotation-filtration reactors using aluminum electrodes. Condition: initial pH 7; filtering velocity, 5.0 m/h.

It is considered that current intensity determines the coagulant dosage, the size and production rate of bubble and the flocs growth [37], which influence the mixing and mass transfer and further the efficiency of color removal. The result was ascribed to the fact that when the current intensity increased, the amount of the aluminum dissolved from the anode increased according to Faraday's law, resulting in more aluminum hydroxides and polyhydroxides for the flocs formation removal of color [34]. Moreover, the increase in current intensity was accompanied with the increase in bubbles density and decrease in bubbles' size [4], resulting in a greater upwards momentum flux. Meanwhile, the attachment step between gas bubbles and pollutants was enhanced, and more pollutants were carried out by gas bubbles [38]. As observed by other researcher, an increase in current yielded an increase in the efficiency of color removal [31]. However, the current was further increased to 2.0 A beyond the optimal value (1.5 A) would generate excessive bubbles, so that there was a strong likelihood for bubbles to coalesce together instead of attachment with pollutants [38,39], causing lower effects for color removal.

In addition, the electrode consumption was also taken into consideration. The amount of aluminum dissolved at different current intensities was estimated by the Faraday's Law:

$$\Delta m = \frac{ItM}{zF} \tag{11}$$

where m is the dissolved aluminum (g), t is the time, z is the number of electrons in the redox reaction, F is Faraday's constant, and M is the molecular weight of Al [18].

The increase in current intensity resulted in the increase in dissolved aluminum, which would be faster than the production of aluminum hydroxides and polyhydroxides. Therefore, a further increase in current did not achieve a higher removal of color. When the current intensity increased to 1.5 A, the increase in current did not achieve higher color removal and led to a relatively high aluminum electrode consumption. Resembling results were exhibited in other researches. Holt et al. [37] ascribed to the supply of aluminum ions is generated rapidly compared to the coagulation process. Kobya et al. [40] reported that decolorization efficiency increased as the increase in current density

Table 5
Analysis of variance (ANOVA) for the removal of DOM

Source	Sum of squares	Degree of freedom	Mean square	*F*-value	*p*-value Prob. > *F*
Model	237.79	9	26.42	41.56	<0.0001
A	3.13	1	3.13	4.92	0.0621
B	50	1	50	78.65	<0.0001
C	21.13	1	21.13	33.23	0.0007
AB	9	1	9	14.16	0.0071
AC	20.25	1	20.25	31.85	0.0008
BC	1	1	1	1.57	0.25
A^2	130.87	1	130.87	205.86	<0.0001
B^2	1.92	1	1.92	3.02	0.1259
C^2	3.6	1	3.6	5.67	0.0488

Notes: R^2: 0.9816, adjusted R^2: 0.9580, predicted R^2: 0.8968, adequate precision: 22.076 and C.V.: 6.39%.

Table 6
Optimization scheme of the experiments

NO.	PFC dosage (mg/L)	Filtering velocity (m/h)	Current intensity (A)	Residual Color (Pt–Co units)	Desirability
1	36.07	5.00	2.00	4.51669	0.762279
2	36.16	5.00	2.00	4.51713	0.762256
3	35.91	5.00	2.00	4.51819	0.762201
4	36.25	5.00	2.00	4.51835	0.762192
5	36.36	5.00	2.00	4.5211	0.762047
6	35.73	5.00	2.00	4.5232	0.761937
7	36.12	5.00	1.99	4.54444	0.760819
8	36.93	5.00	2.00	4.55745	0.760134
9	36.31	5.00	1.99	4.568	0.759579
10	34.90	5.00	2.00	4.59439	0.75819
11	36.09	5.00	1.98	4.61383	0.757167
12	37.46	5.00	2.00	4.62314	0.756677
13	34.49	5.00	2.00	4.65655	0.754919
14	35.33	5.00	1.98	4.67129	0.754142
15	36.32	5.02	2.00	4.675	0.753947
16	33.86	5.00	2.00	4.7918	0.7478
17	38.36	5.00	2.00	4.80629	0.747037
18	36.25	5.00	1.90	5.03316	0.735097
19	36.90	5.00	1.87	5.16828	0.727985
20	37.37	5.00	1.83	5.38701	0.716473
21	40.11	5.00	0.50	7.62438	0.598717
22	39.32	5.00	0.80	7.73918	0.592675
23	39.47	5.00	0.75	7.74644	0.592292

and the decolorization efficiency did not change significantly, as the applied current density increased from 100 to 250 A/m^2. And Gao et al. [34] found that the higher the current density was, the more rapid the ECF treatment for algae removal would be.

3.6. Optimization of the reactor using response surface methodology (RSM) approach

RSM is a multivariate factorial design which will allow a statistical assessment of the results and statistical support of the conclusions. The interaction of the factors was evaluated based on the establishment of a continuous variable surface model through RSM. And relatively less experimental groups are needed, which saves the time and resource. In consideration of the interaction of various factors, the effects of PFC dosage, filtering velocity, and current intensity were investigated using RSM with aluminum electrode. Based on the results above, the appropriate boundary value of every factor has been determined to proceed to the Box–Behnken design orthogonal experiments of three factors and three levels (Table 3). Table 5 shows the analysis of variance (ANOVA) of the model. The model was significant ($p < 0.0001$) with F-value of 41.56. The coefficient of determination (R^2) for color was observed to be 0.9816. The closer the R^2 is to unity, the stronger the model and the better it predicts the response. The predicted multiple correlation coefficient (predicted R^2) value of 0.8968 was in a reasonable agreement with the adjusted multiple correlation coefficient (adjusted R^2) value of 0.9580. The coefficient of variance (C.V.) of this model was reported as 6.39%. The lower the C.V. value, the greater is the precision and reliability of the experiments carried out. Adequate precision measures the signal to noise ratio and a ratio greater than 4 is desirable. The ratio of 22.076 indicates an adequate signal.

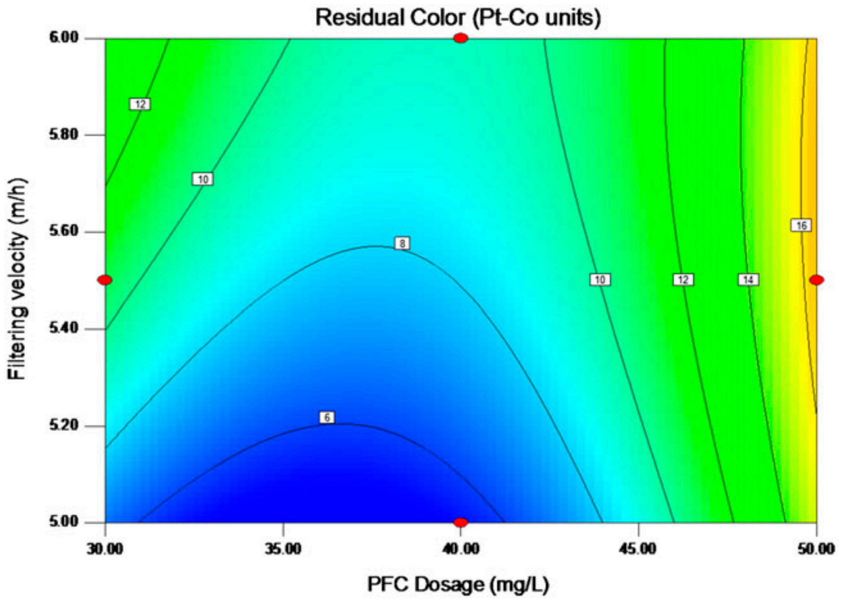

Fig. 6. The optimized contours between the PFC dosage and current intensity for residual color.

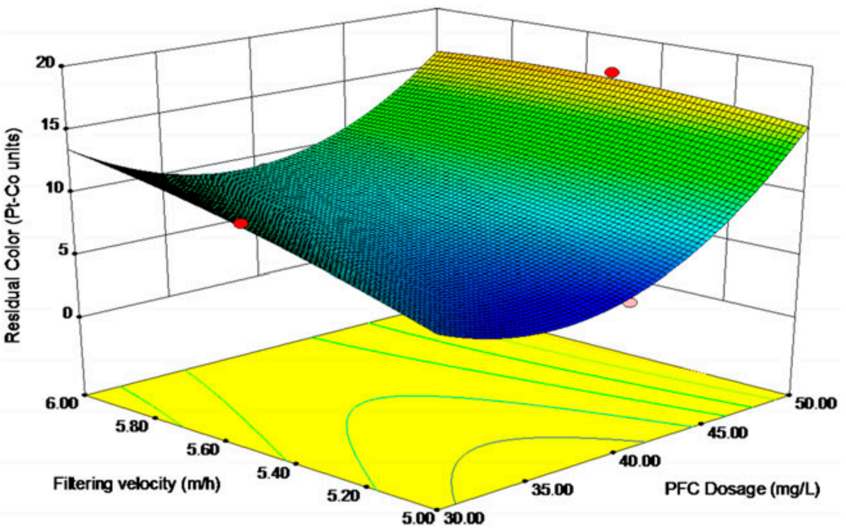

Fig. 7. The optimized response surface between the PFC dosage and current intensity for residual color.

The modified cubic model describing the correlation between the variables and the residual color was shown as follows:

$$\begin{aligned}\text{Residual color} = &-71.9 - 3.12A + 48.37B - 2.72C \\ &- 0.30AB + 0.30AC - 1.33BC + 0.06A^2 \\ &- 2.70B^2 - 1.64C^2\end{aligned} \tag{12}$$

Based on the experimental results and the fitting model analysis, the optimization for the level of every factor could be conducted on the basis of the minimum residual color (Table 6). When the filtering velocity was 5.00 m/h, the PFC dosage was 36.07 mg/L, and current intensity was 2.00 A, the minimum residual color of 4.52 Pt–Co units could be obtained. The optimized contours and response surface between the PFC dosage and current intensity for the residual color of the No. 1 optimization scheme are shown in Figs. 6 and 7, respectively, under the conditions that the filtering velocity was 5.00 m/h and electrode material was aluminum.

4. Conclusions

In this study, both the jar test coagulation and the pilot scale continuous flow electrocoagulation/electroflotation-filtration process were successfully carried out to removal the color from groundwater. The effects of various electrocoagulation/electroflotation-filtration reactor's operational parameters were researched.

It was found that PFC was a relatively efficient coagulant and aluminum electrode was better. Increase in current intensity caused a increase in color removal efficiency and increasing filtering velocity resulted in higher color removal efficiency. Furthermore, the optimization and modeling of residual color in groundwater were conducted using RSM with BBD. By using RSM the optimum operation conditions were obtained (filtering velocity of 5.00 m/h, PFC dosage of 36.07 mg/L, and current intensity of 2.00 A with aluminum electrode) and the minimum residual color was 4.52 Pt–Co units.

The results of this study indicated that EFC combined with filtration process was a hopeful technique to treat groundwater with high chroma and low turbidity causing by DOM and other matters. This study can be used for reference for further application of EFC combined filtration continuous flow reactors to treat groundwater with high chroma and low turbidity.

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