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Taguchi experimental design for electrocoagulation process using alternating and direct current on fluoride removal from water

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ABSTRACT

Taguchi experimental design with the alternating and direct current (AC and DC) electrocoagulation (EC) using initial fluoride concentration of contaminant, retention time, pH, electrode type, and voltage was used on fluoride removal. DC current is significantly more efficient than AC, and Al has a higher performance than Fe electrode ($p < 0.05$). Neutral pH is more effective than acidic and alkaline condition in which significant differences are observed in reaction time of 60 min ($p = 0.02$). Elevation of cell potential from 15 to 25 V led to enhancing efficiency, but beyond these range fluoride removal declined. Fluoride concentration increase from 3 to 8 mg/l declined the EC performance with a significant impact in reaction times of 20 and 40 min ($p = 0.004$). In similar cell potential, increasing in reaction time from 20 to 60 min led to decreasing in residual fluoride from 1.75 to 1.06 mg/l, and 1.64 to 0.76 mg/l in 25 and 40 V, respectively.

Keywords: Taguchi experimental design; Fluoride; Electrocoagulation; Alternating and direct current

1. Introduction

Water is one of the basic requirements for human health. Globalization, uncontrolled population, and other factors are creating safe drinking water shortage; therefore, the issue is a worldwide concern. Fluoride is the 13th most abundant element in the earth's crust with an average concentration of 0.3 mg/kg [1,2]. The beneficial and intoxicative detrimental manifestation effects of fluoride on human health are dependent on its intake level [3].

High concentration of fluoride is a persistent and nondegradable inorganic contaminant that can be enriched in water resources through a combination of anthropogenic activities including indiscriminate discharge of industrial effluents, specially electroplating, glass, steel, ceramic, and fertilizer industries, as well as

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natural processes. In the latter, unknown geochemical processes have contaminated the ground water with fluoride in many countries [4–6]. In Iran, the problem of high fluoride level was considerable in seven province ground waters which depicts the 23% of the provinces [2]. WHO set the permissible level of fluoride as 1.5 mg/l, and when the concentration of fluoride is above this limit, cosmetic, and systemic disorders including dental and skeletal fluorosis and lesions of the endocrine glands, thyroid, liver, decrease growth and intelligence, paralysis and cancer, neurological disorder, kidney damage, and infertility are arising in exposed population [7,8]. Although, many of the disruptive health effects relevant to high levels of fluoride intake via drinking water as a major source of fluoride intoxication in human body are able to be eliminated, the dental and skeletal fluorosis as traditional symptoms of chronic exposure to fluoride is irreversible and no treatment exists. Therefore, keeping the fluoride intake within the safe limits is the only remedy [2,5]. Due to these detrimental effects, water pollution by fluoride has been considered as legitimated public health concern worldwide [5]. Therefore, development of innovative, more effective, and inexpensive techniques for the elimination of fluoride from water is an urgent need. Several techniques including nanofiltration and electrodialysis [9], membrane processes [10], adsorption [11], and chemical precipitation and coagulation [12] were used to prevent public health risks and to reduce the prevalence of fluoride-related disorders in endemic areas. Some of these techniques have serious drawbacks in terms of high cost, techno-feasibility, simplicity, generation of secondary pollutants, and large volumes of sludge or wastes which pose serious environmental issues [10,13].

In recent years, a host of environmental friendly and promising technique based on electrochemical technology known as electrocoagulation (EC) is being developed and existing ones have improved with relatively low operating cost, applicability for the removal of wide range of organic and inorganic pollutants such as pathogens, not requiring chemical additions, less detention time, and rapid settleability of electro-generated flocs [14,15]. In this process (EC), the coagulant is generated *in situ* by electrolytic oxidation of an appropriate anode material as a sacrificial electrode [16]. So, this promising technique offers an alternative for use of coagulants and co-coagulants including metal salts, polymers, and polyelectrolyte addition for pollutants elimination. Therefore, it can be suggested that the EC process is cost effective and safe technique from points of transportation, storage, and application of coagulants; on the other hand, it can be categorized as a clean process that can be operated with renewable energy resources including solar irradiation and windmills [15]. Although, both direct and alternating current (DC and AC) can be applied in EC technique, but formation of an impermeable layer on anode and corrosion of cathode are the most drawbacks of DC current which may lead to process performance decline. Therefore, in recent years overcoming of DC disadvantages has been considered by adopting an AC application as an alternative in EC process [17,18].

Since, fluoride is considered as a cosmetic and systemic disruptive pollutant worldwide, and controversial ideas are being considered for AC and DC application from points of operation and performance views; this study illustrates the EC process efficacy via DC and AC current for the removal of excess fluoride from water.

2. Material and methods

A batch pilot-scale EC reactor with electrodes connected to the EC reactor with monopolar parallel connection mode with two anodes and two cathodes electrodes was designed and constructed for fluoride

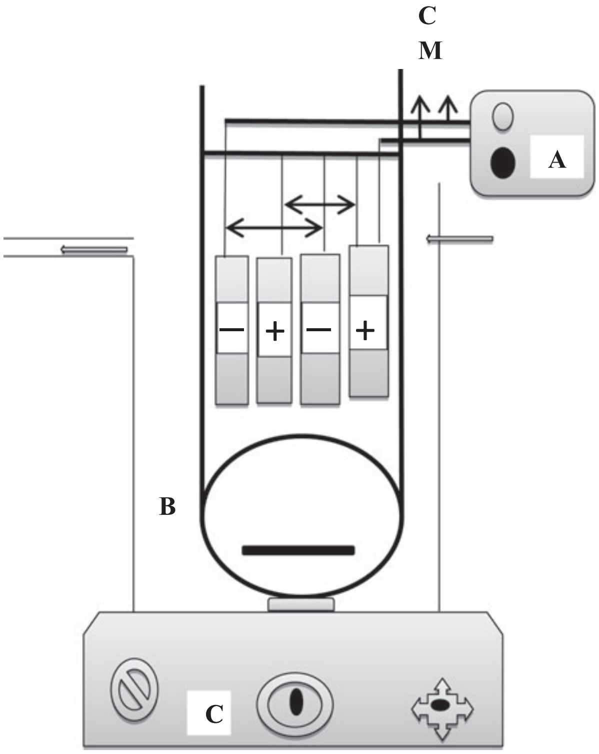

Fig. 1. Schematic setup of electrocoagulation system.
Note: A: power supply, B: thermostatically water, CM: current monitoring; C: magnetic stirrer.

removal (Fig. 1). For the electrochemical cell, four aluminum or iron plates (dimension 110 mm × 110 mm × 2 mm) were used as anodes and cathodes electrodes. The electrodes with total surface area 0.726 dm^2 were dipped 140 mm into an aqueous solution (net volume 2 l) in a Plexiglas vessel (Øint = 14 cm, *H* = 30 cm). The distances between the two conterminous electrode plates were kept constant at 10 mm during the experiments. All experiments were performed within high density Plexiglas container at room temperature (20 ± 2˚C), and the electrolyte temperatures were controlled at the desired values with a variation of ±2˚C by adjusting the flow rate of thermostatically controlled water through an external glass container. Alternating or direct current from a power supply (TDGC2-200VA, 0–250 V) was passed (15, 25 and 40 V) through the solution via the electrodes. A digital multimeter power displayer (DT 700D, AKB) was used to monitor the passed voltage and current. Conversion of AC to DC current was done via four inverter diodes. All of chemicals and reagents were of analytical grade (Sigma-Aldrich) and used without further purification. Fluoride stock solution was prepared by dissolving 221 mg anhydrous sodium fluoride (NaF) in 1,000 ml distilled water in volumetric flask and desired concentrations (3, 5, 8 mg/l F) were prepared by dispending of an appropriate aliquots to 2,000 ml distillated water and used as electrolyte solution. The electrolyte pH was adjusted (3, 7, and 10) with HCl or NaOH (0.1 M) and measured with a calibrated pH meter (METTLER TOLEDO, GmbH, Seven Easy). After electrolyte preparation with the appropriate fluoride concentration and pH adjustment, the EC process with AC or DC current was started while complete mixing of the reactor constituent was done via magnetic bar stirrer (Cole-Parmer, ST11, Dia 5 × 15 mm). At the end of the experimental runs (20, 40, and 60 min) the aliquot of samples (100 ml) were taken for fluoride measurement by centrifugation on 10 min and 1,500 RPM (Eppendorf, 5810 R) and filtration with 0.45 μm membrane.

Fluoride measurement was conducted based on spectrophotometric technique on 570 nm (Visible-Cecil-1011) with SPANDS method according to the instruction described in standard methods for the examination of water and wastewater in which Al^{+3} interferences were controlled by buffer addition [19].

EC efficiency, expressed as percent EC was calculated using the following equation:

$$\%\text{EC removal} = \frac{(C_0 - C_{\text{Res}}) \times 100}{C_0} \tag{1}$$

where C_0 and C_{Res} are the initial and residual fluoride concentration (mg/L) in solution, respectively.

To yield a balanced design, an orthogonal array is constructed based on five operational variables [pH, F concentration (mg/l), voltage (V), reaction time (min), and electrode type (Al or Fe)] with three levels and combinations in equal number of experiments for AC and DC currents, separately. Taguchi approach based on the analysis of loss function was used to relent of the signal (product quality)-to-noise (uncontrollable factors) ratio, and is applied as the enlightening parameter to affirm the quality of experiment together with the validity of the result (response). The S/N ratio, in fact, conceives the fluctuation or level of exactitude of each response for each single experiment (trial) in the total set (replication). Calculation of the signal-to-noise ratio depends on the experimental objective; therefore, three categories of S/N ratio analysis viz. "Maximum-is-better," "Nominal is best," and "Less-is-better" are available [10]. As the present study constraints minimum residual fluoride by EC process, the S/N ratio was determined based on LIB approach with the following Eq. (2):

$$\frac{S}{N} = -10 \log\left[\sum \frac{y^2}{n}\right] \tag{2}$$

where n is the number of observations, and y is the observed response (fluoride residual concentration). The constructed orthogonal array [L18 (35)] based on five independent variables, viz., A: electrode composition (Al and Fe), B: pH (3, 7, and10), C: voltage (15, 25, and 40 V), D: reaction time (20, 40, and 60 min), and E: initial F concentration (3, 5, and 8 mg/l), each was taken at three levels (except electrode type) for AC and DC current, separately. Only 18 experiments are required to design the simultaneous impact of operational variables. Each experiment was triplicated (1st run, 2nd run, and 3rd run), and the S/N ratio was calculated using Minitab software (version 17). For comparison of the two AC and DC alternatives the experiments for AC and DC currents were performed, separately. Minitab (Ver. 17) and SPSS (Ver. 15) software was used for prediction of the variable influences on response, the fluoride removal percent/residual fluoride, and data analysis, respectively. Normal distribution of data was determined by Kolmogorov–Smirnov test. Descriptive statistics were used for description of data and the independent *T*-test, Mann–Whitney *U* test, one-way ANOVA, Kruskal–Wallis, BonFerroni, and Tukey *Post Hoc* tests were used for validation and quantitation of the parameters effects using statistical package software (SPSS 15) in which statistical results were interrupted at the level of significance $p < 0.05$. All statistical analysis was performed with the residual fluoride concentration; otherwise, it is stated in the text.

3. Results and discussion

All of the experiments for comparison and precision of AC and DC current efficacy for the removal of fluoride conducted separately. Hence, the response values (*F* residual concentration), as well as the *S*/*N* ratio replying to the orthogonal array [L18 (35)] calculated for AC and DC currents, separately (data not shown). The mean of *S*/*N* ratio was analyzed and the optimum level of operational variables was determined from the highest value. The calculated Δ*s* (max–min) are implying identified ranks or influences of the operational variables for fluoride removal or residual fluoride by AC and DC current are presented in Tables 1 and 2, respectively. Comparison of these ranks demonstrates that in AC current, the pH of solution exhibits the highest influence on fluoride removal (Table 1). Similar results were obtained based on the mean and standard deviation of residual fluoride concentration (data not shown). The optimum status was found to be A1-B1-C3-D3-E1 with the corresponding to Al electrode, pH 7, Vol = 40 V, reaction time 60 min, and the initial fluoride concentration of 3 mg/l. As demonstrated in Table 1, in AC current, pH, and initial concentration of fluoride is the predominant parameter for the removal of fluoride.

In Table 2, the influence of variables on residual fluoride in DC current is demonstrated. Therefore, in DC current, the composition of electrode corresponding to Al has the highest effect on fluoride removal. So, the optimum condition was found to be A1-B1-C2-D3-E1 with corresponding to Al electrode, pH 7, Vol = 40 V, reaction time 60 min, and initial fluoride concentration of 3 mg/l. These results imply that in DC current, unlike AC current, the composition of electrode and voltage is the main effective parameter. Comparison of the means of residual fluoride concentration in AC and DC currents revealed that the latter current has the higher efficacy; therefore, in DC current the residual fluoride concentration is lower than the WHO recommended guideline.

In recent years, formation of impermeable layer in DC's current anode has been considered as a drawback so, adaption of AC current was considered as an alternative in EC technique. Moreover, we evaluated the effects of current type (AC and DC) as an independent variables or operational parameters via statistical analysis for the removal of fluoride. Analysis of data via independent *T*-test and Mann–Whitney *U* test show that in different reaction times (20, 40, and 60 min) the median and mean of the residual fluoride in AC and DC current has significant differences; hence, in the latter condition the residual fluoride is significantly lower than the AC current ($p < 0.05$) (Table 3). As demonstrated in Table 3, the DC current is more effective than the AC current and WHO maximum permissible level via latter current can be provide only in 60 min reaction time. But, DC current provided WHO guideline value in all of reaction times. Based on this result, association of the other variables effects on water defluoridation was followed by DC condition. Although, Vasudevan et al. [17,20] reported that application of AC current is efficient than DC current but, they did not conduct statistical analysis to reveal the effectual differences for this claimed alternative. On the other hand, in fluoride and iron removal the differences between these currents are 1.25 and 0.28%, respectively, by AC current which may not be considered as statistically significant. In the present study, statistical analysis implied that DC current can play imperious role in EC process efficacy.

Therefore, it may not be concluded that AC current is better than DC from point of process performance in pollutant removal. Although, formation of impermeable layer in AC current may lead to process deficiency, but this limitation can be overcome by agitation as observed by the authors.

Table 1
S/*N* ratios means in AC current

Level	A (Electrode)	B (pH)	C (Voltage)	D (Time)	E (C_0)
1	−3.607*	−2.511*	−5.275	−5.275	−2.054*
2	−4.777	−6.238	−4.296	−4.895	−5.087
3	–	−3.829	−2.748*	−2.847*	−5.386
Δ	1.170	3.727	2.527	2.428	3.332
Rank*	5	1	3	4	2

*The maximum mean *S*/*N* ratio implies the optimum condition.

Table 2
S/N ratios means in DC current

Level	A (Electrode)	B (pH)	C (Voltage)	D (Time)	E (C_0)
1	4.88232*	0.31641*	−2.02869	−0.66343	0.71758*
2	−4.54164	0.21976	1.89605*	−0.64979	−0.13666
3	–	−0.02515	0.39319	1.82424*	−0.07944
Δ	9.42396	0.34156	3.92474	2.48767	0.85424
Rank	1	5	2	3	4

*The maximum mean S/N ratio implies the optimum condition.

Table 3
Assessment of AC and DC current on residual fluoride

	Residual fluoride (time 20 min), Mann–Whitney *U* test					
Current	Min	Max	Ave.(CI %95)	SD	Median	*p*
AC ($n = 18$)	0.72	0.32	1.83(1.58–2.08)	0.498	1.99	<0.001*
DC ($n = 18$)	0.31	1.97	1.21(0.92–1.5)	0.583	1.37	
	Residual fluoride (time 40 min), independent *T*-test					
Current	Min	Max	Ave.(CI %95)	SD		*p*
AC ($n = 18$)	0.37	2.7	1.82(1.52–2.12)	0.611		0.008*
DC ($n = 18$)	0.16	2.28	1.19(0.83–1.55)	0.729		
	Residual fluoride (time 60 min), Mann–Whitney *U* test					
Current	Min	Max	Ave.(CI %95)	SD	Median	*p*
AC ($n = 18$)	0.61	2.2	1.46(1.2–1.72)	0.52	1.46	0.021*
DC ($n = 18$)	0.14	1.97	1(0.68–1.32)	0.64	0.86	

*p values < 0.05 are significant, initial fluoride concentration = 8 mg/l.

As demonstrated in Tables 1 and 2, the composition of the electrode has different effect in AC and DC current; hence, in DC current the electrode composition has the predominant effect, but in AC current the electrode has the least impact on fluoride removal. These findings were validated via statistical analysis. As shown in Table 4, the median and mean of the residual fluoride is significantly different in Al and Fe electrode material; therefore in the latter electrode the residual fluoride is higher than the Al and greater than the WHO permissible level, revealing the less efficacy of Fe electrode material. Also, Khandegar and Sahora reported the higher efficiency (79.68%) for Al–Al than Fe–Fe (56%) electrode material which is compatible with the findings of the present study [15].

The present study revealed that, the electrode materials can influence the fluoride removal via EC process and Al has a high efficacy when used as an electrode. This may be relevant to F–Al and F–Fe affinity. Thus, in Pauling scale the differences between F–Al electronegativity (2.37) are higher than F–Fe (2.15), which implies that the F–Al affinity is higher than F–Fe. Similar result considering the electrode material effects was reported by Lacasa et al. [21], in which the effects of Al and Fe electrode composition were discussed for nitrate removal. It can be concluded that the efficacy of electrode composition in the EC deponents on both composition of the removed contaminants and the operational condition.

As demonstrated in Tables 1 and 2, the pH of solution has different role in AC and DC current; therefore, in AC current this variable has the highest influence, but acts unlikely in DC. Since, pH is an important parameter for the chemical reactions performance, the analysis of the pH effects and differences of the mean and median residual fluoride concentration is necessary. Analysis of data with Kruskal–Wallis test implied that in all of the reaction times the median of residual

Table 4
Analysis of residual fluoride in accordance with electrode composition

	Residual fluoride (time 20 min), Mann–Whitney *U* test					
Electrode	Min	Max	Ave.(CI %95)	SD	Median	*p*
Al ($n = 18$)	0.31	2.25	1.21(0.86–1.55)	0.692	1.02	<0.016*
Fe ($n = 18$)	0.84	2.3	1.83(1.66–1.99)	0.331	1.79	

	Residual fluoride (time 40 min), independent *T*-test				
Electrode	Min	Max	Ave.(CI %95)	SD	*p*
Al ($n = 18$)	0.16	2.21	1.2(0.86–1.55)	0.71	0.01*
Fe ($n = 18$)	0.37	2.7	1.83(1.5–2.12)	0.64	

	Residual fluoride (time 60 min), independent *T*-test				
Electrode	Min	Max	Ave.(CI %95)	SD	*p*
Al ($n = 18$)	0.14	1.82	0.85(0.6–1.1)	0.525	0.001*
Fe ($n = 18$)	0.61	2.2	1.6(1.4–1.8)	0.47	

*p values < 0.05 are significant, initial fluoride concentration = 8 mg/l.

fluoride in pH 7 is lower than the pH 3 and 10, in which pH 7 has the least concentration. The differences of the median in reaction times of 20 and 40 min are not significant, but in reaction time of 60 min, the median differences are significantly different ($p = 0.02$). The validation of the pH differences was conducted by Man-Whitney test and modification of p values and multi-comparison by Bonferroni shows that the differences between various pH of solution in reaction time of 60 min are relevant to differences of pH 7 and 10; thus the p value of two points of pH (7 and 10) is 0.039, which is statistically significant. These findings imply that fluoride removal with EC is dependent to pH with forward–backward effects; therefore elevation of pH from 3 to 7 had led to enhancing of fluoride removal, but beyond this point the residual fluoride increased in the treated water and defluoridation decreased. This phenomenon is relevant to $Al(OH)_3$ amphoteric behavior and influenced by the speciation of Al in various pH; hence, the alkaline and acidic surrounding pH induces the formation of the soluble monomeric anions ($Al(OH)_4^-$) and Al^{+3}, respectively; which are not useful in fluoride removal [20,22].

The performance of EC process influenced by applied cell potential and electrolysis time has impact on the pollutant removal via coagulant generation and rate and size of hydrogen bubble production [23]. The influences of electrolysis reaction time on residual fluoride are shown in Fig. 2. The result implied that elevation of reaction time led to decreasing of residual fluoride which complies by coagulant generation in EC reactor. This phenomenon can be discussed based on Faraday's law; therefore, increasing in reaction time coordinate the Al^{+3} concentrations and their hydroxide flocs and consequently the process relent is improved [24].

Increasing in voltage from 15 to 25 and 40 V led to enhancing of fluoride removal from 76 to 85.7 and 90.6%, respectively. Therefore, after 60 min reaction time, residual fluoride is 1.92 mg/l for 15 V; hence, for 25 and 40 V it is 1.14 and 0.75 mg/l, respectively. This can be relevant to sufficient $Al(OH)_{3-x}F_x$ formation as coagulant [23,25]. Similar result was observed in

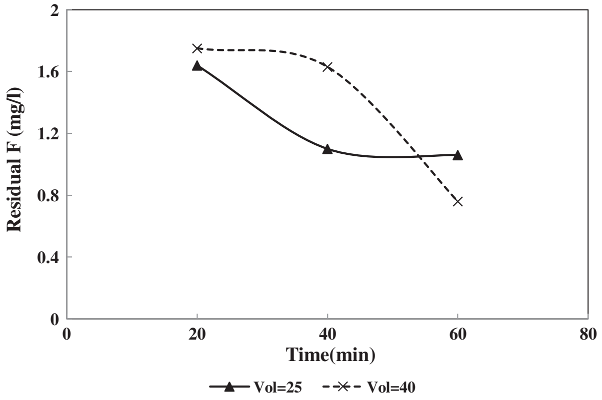

Fig. 2. Effect of electrolysis time on fluoride residual.

40 min reaction time and cell potential of 15 and 25 V, so cell potential increasing in this range leads to increasing in fluoride elimination from 72.9 to 84.5%, respectively; but beyond 25 V, the defluoridation was declined to 77.5%. Similar results were observed in 20 min reaction time; therefore, elevation of cell potential from 15 to 25 and 40 V led to decreasing of EC performance from 87.4 to 78.4 and 76.4%, respectively. Since, coagulant production and floc flotation by H_2 bubbles simultaneously influenced the EC performance; this phenomenon may be relevant to high agitation and breaking of the generated flocs or declining of H_2 gas floatation effects which leads to decreasing of fluoride removal in which generated coagulant was not optimally consumed. Similar results was reported by Ifill and Etsell for deductive agitation effects in EC process [26]. In addition, Khatibikamal et al. [23] reported that operation of EC process with the higher current densities may not lead to enhanced pollutant removal. Hence, it can be concluded that in EC process, balancing of the cell potential in allowable ranges, agitation speed and retention time is necessary for optimized production/consumption of coagulant and H_2 gas bubbles are being the efficient mode of operation. Examination of the appraised voltages and reaction times implied that the median of residual fluoride has no significant differences in surveyed voltages and reaction time of 20 min, but have significant differences in reaction time of 40 and 60 min (p = 0.038 and 0.001, respectively). Comparison of the residual fluoride median in reaction times of 40 and 60 min and various voltages revealed the

Table 5
Effect of pH on fluoride removal

	Residual fluoride (reaction time: 20 min), 25 V			
pH	Ave.(CI %95)	SD	Median	p (Kruskal–Wallis test)
3	1.65(1.24–2.1)	0.721	1.95	0.151
7	1.25(0.9–1.62)	0.58	1.3	
10	1.65(1.32–1.97)	0.454	1.76	
pH	Residual fluoride (reaction time: 40 min), 25 V			
	Ave.(CI %95)	SD	Median	p (Kruskal–Wallis test)
3	1.75(1–2.24)	0.732	2.06	0.1
7	1.18(0.81–1.55)	0.61	1.4	
10	1.63(1.12–2.13)	0.795	1.72	
pH	Residual fluoride (reaction time: 60 min), 25 V			
	Ave.(CI %95)	SD	Median	p (Kruskal–Wallis test)
3	1.36(1–1.73)	0.577	1.47	0.02*
7	0.85(58–1.12)	0.43	0.92	
10	1.5(1.04–1.92)	0.7	1.75	

*p values < 0.05 are significant.

Table 6
Association of initial fluoride concentration on EC defluoridation

	Residual fluoride (reaction time: 20 min), 25 V			
Fluoride Con. (mg/l)	Ave.(CI %95)	SD	Median	p (Kruskal–Wallis test)
3	0.95(0.54–1.36)	0.65	0.75	0.004*
5	1.75(1.53–1.97)	0.34	1.74	
8	1.85(1.6–2.1)	0.4	1.9	
	Residual fluoride (reaction time: 40 min), 25 V			
Fluoride Con. (mg/l)	Ave.(CI %95)	SD		p (ONE WAY ANOVA)
3	1.1(0.73–1.5)	0.6		0.004*
5	1.42(0.52–2.3)	0.85		
8	1.8(1.46–2.14)	0.68		
	Residual fluoride (reaction time: 60 min), 25 V			
Fluoride Con. (mg/l)	Ave.(CI %95)	SD	Median	p (Kruskal–Wallis test)
3	0.97(0.61–1.33)	0.57	0.9	0.107
5	1.22(0.78–1.37)	0.7	1.3	
8	1.5(1.16–1.84)	0.53	1.54	

*p values < 0.05 are significant.

significant differences ($p = 0.038$ and <0.001, respectively) and these differences were validated by Man-Whitney and Bonferroni technique. Pair comparison of these statistical differences in reaction time of 40 min revealed that the differences of 25 and 40 V, 15 and 40 is not significant ($p = 0.15$ and $p = 1$, respectively), but in 15 and 25 V is 0.072, which is significant ($\alpha = 0.1$). These results imply that the statistical differences of residual fluoride are relevant to 15 and 25 V. In reaction time of 60 min the pair comparison of differences demonstrated that the differences are originated from the 15 and 25 and 25 and 40 V (Table 5).

Initial concentration of pollutant is another variable that has a key role in pollutant remediation from aqueous solution via EC. In order to find out the influences of initial concentration of fluoride on defluoridation performance, the experiments were done by altering initial fluoride concentration from 3 to 8 mg/l at the same cell potential and reaction time. The result implied that elevation of fluoride concentration from 3 to 8 mg/l led to decreasing in EC performance (Table 6). As demonstrated in Table 6, in reaction times of 20 and 40 min, augmentation of fluoride concentration from 3 to 8 mg/l led to elevation of residual fluoride concentration to a level higher than the WHO permissible guideline value; but, in reaction time of 60 min for all concentrations, the WHO guideline value was met. Since, in EC process the highly charged polymeric metal hydroxide species accessibility as electrocoagulant has a key role in process efficiency, the decreasing in EC performance or residual fluoride elevation can be attributed to insufficient generation of coagulant or perturbation in coagulant and pollutant ratios. Similar results were reported in previous studies [15,27].

In the present study, the comparison of the median residual fluoride in appraised concentration and reaction time revealed that in retention time of 60 min the fluctuation of initial fluoride concentration had not led to significant differences on the average of residual fluoride ($p = 0.107$); but, in retention time of 20 and 40 the significant effect was observed ($p = 0.004$). Pair comparison of medians in reaction time of 20 min confirmed that the difference of 5 and 8 mg/l is not significant ($p = 1$), but significant differences were observed in 3, 5 and 3, 8 mg/l of fluoride concentrations ($p = 0.12$ and 0.021; $\alpha = 0.1$ and 0.05, respectively); thus in this detention time, the differences are relevant to these concentrations. In reaction time of 40 min, the validation of differences via Tukey *Post Hoc* test showed that concentrations of 3 and 8 mg/l have led to statistical differences.

4. Conclusion

Taguchi experimental design is an appropriate alternative versus conventional statistical for sample size that can lead to resources save in experimental researches. Based on statistical analysis, DC current has a higher efficacy than AC current and the composition of cell material is important in pollutant removal which is dependent to affinity of pollutant and generated coagulant; consequently, Al cell is better than Fe in fluoride removal. Due to amphoteric characteristics of Al, the neutral pH of surrounding is appropriate for fluoride removal and the least residual concentration can be obtained in neutral pH. Operation of EC with the higher cell potential may not lead to enhanced pollutant removal; so, balancing of cell potential and reaction time with the other operational parameters is necessary. Increasing in the initial concentration of contaminant is another variable that influences the EC performance with unbalancing of electrogenerated coagulant and pollutant mass ratios that should be considered in EC operation.

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