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DEFLUORIDATION OF POTABLE WATER USING ELECTROCOAGULATION

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ABSTRACT
The presence of fluoride in drinking water is a global concern stemming from both natural and industrial sources, potentially leading to health issues like fluorosis with prolonged exposure. The World Health Organization sets a standard of 1.5 mg/l for fluoride in drinking water.
To address this, researchers conducted batch experiments using electrocoagulation (EC), an electro-chemical technique that removes suspended and dissolved particles through electrolysis. They explored the impact of four key factors: current density (13-65A/m2), inter-electrode distance, initial fluoride concentration, and reaction time on fluoride removal, while also calculating the specific energy consumed.
Using Design Expert 8.0.7.1, a full 24 factorial design was employed to optimize fluoride removal from drinking water. Different levels of inter-electrode distance (20 and 40 mm), current density (13 and 65A/m2), initial fluoride concentration (4 and 6 mg/L), and reaction time (10 and 50 minutes) were examined. A regression model equation was developed and validated with a high R2 value from ANOVA. The optimization aimed for residual fluoride concentration below 1 mg/l and minimized specific energy consumption.

Keywords: Defluoridation, Electrocoagulation, Fluorosis, Fluoride Removal, Inter Electrode, ANOVA

1. INTRODUCTION
In the halogens fluorine is the lightest member and is highly reactive. It therefore does not exist as elemental fluorine. It has high electronegativity and thus has a strong affinity to gain a negative charge and thus exist in form of fluoride ion. It is the 13th most abundant element on earth.[1,2]It is found mainly as sodium fluoride or hydrogen fluoride chiefly associated with minerals such as fluorspar ,cryolite etc. The percentage of the fluorine content present in these minerals is shown in Table 1.

S.NO	Mineral with its chemical formula	% fluorine
1	Sellaite , MgF2	61%
2	Villianmite, NaF	55%
3	Fluorite (Fluorspar) ,CaF2	49%
4	Cryolite, Na3AlF6	45%
5	Bastnaesite, (Ce,La) (CO3)F	9%
6	Fluorapatite , Ca3(PO4)3F	3-4%

Table 1: Minerals and their fluoride content

1.1Objectives of the Study
The present study is carried to understand the main effects and interaction effects of four operating parameters: current density, inter electrode distance, initial fluoride concentration, and reaction time on residual fluoride and specific energy consumed.
A batch reactor study was conducted and the experimental results so obtained were analyzed using Design Expert 8.0.7.1.A full 2^4 factorial design of experiment was undertaken to examine the main factors affecting the fluoride removal and their interactions. Two levels for each factor were used; inter electrode distance (20 and 40 mm), current density (13 and 65A/m^2), and initial fluoride concentration

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(4and 6 mg/L) and reaction time (10 and 50 minute). A regression equation was developed which was validated by high R^2 value of ANOVA. Optimization was targeted at residual fluoride concentration less than 1 mg/l and minimum specific energy consumption

2. REVIEW OF LITERATURE

2.1. SOURCES OF FLUORIDE FOR HUMAN EXPOSURE

Main sources of fluoride for human exposure are Water, Air, Food and beverages other than water, dental products, drugs etc.

2.1.1 WATER

Drinking water is typically the largest single contributor to daily fluoride intake [18]. For a given individual, fluoride exposure (mg/kg of body weight per day) via drinking-water is determined by the fluoride level in the water and the daily water consumption (litres per day).

2.1.2 AIR

Fluorides are widely distributed in the atmosphere due to dust, industrial production of phosphate fertilizers, coal ash from the burning of coal and volcanic activity.

2.1.3 DENTAL PRODUCTS

A number of products used by children to reduce tooth decay contain fluoride. This includes toothpastes and other supplements. Different products add to different degrees of fluoride. [18]

2.1.4 FOOD AND BEVERAGES OTHER THAN WATER

Vegetable, milk and fruits generally have low fluoride content and thus exposure is typically less but high fluoride content has been found to be associated with barley and rice [18].

2.1.5 DRUGS

Prolonged use of certain drugs has been associated with the chronic adverse effects of fluoride e.g. sodium fluoride for treatment of osteoporosis, niflumic acid for the treatment of rheumatoid arthritis, use of fluoride mouth rinses (Profolo) to render the tooth stronger.

2.1.6 INHERITED

It has now been confirmed that fluoride is inherited from mother to a newborn by passing through the placenta and thus causes skeletal fluorosis in the foetus [20]. Further it leads to dissolution of calcium from the tissues thus weakening them.

2.2. METHODS TO REMOVE FLUORIDE FROM WATER.

Because it is costly and difficult to remove, most communities faced with high F concentrations in their source water first seek an alternate source of supply before considering treatment. If an alternate source is not available, many different methods of defluoridation are available. Several processes have been proposed for removing F from drinking water including [25]:

1. Coagulation and precipitation,
2. Selective adsorption,
3. Ion Exchange,
4. Membrane processes,
5. Electrochemical method

3. MATERIALS AND METHODS

3.1. ELCTROCOAGULATION APPARATUS

A batch defluoridation apparatus consist of a beaker with an effective volume of 2 liters containing two aluminium electrodes as shown in the figure. Each aluminium electrode having a purity of 98.99% and having dimension (74mm×67mm×1mm) and (84mm×71mm×2.5mm) were used as anode and cathode respectively. The effective area of anode used for electrocoagulation is 0.0114 m2. The electrodes were connected in monopolar configuration to a D.C power supply (Testronix, 230V DC).To achieve uniform mixing, a magnetic stirrer is used.

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Fig. 1. Diagram of the experimental setup

3.2. ANALYTICAL PROCEDURE

All the chemical or reagents prepared and physiochemical parameter analysis were according to the methods as described in the "Standard Methods for examination of water and waste water [31].

3.3. SAMPLE ANALYSIS

To study the effect of EC process on drinking water for defluoridation, samples were prepared by dissolving desired amount of sodium fluoride and sodium chloride (2mM) in tap water of MUIT, Lucknow. The tap water quality of MUIT, Lucknow is shown in Table 1

Chemical Parameter	Concentration
Alkalinity (total), ppm	250
Hardness as CaCO3, ppm	180
Total dissolved solids, ppm	100
Chloride, ppm	45.07
Conductivity, mS/cm	0.3
Ph	7.81

Table 2. Water quality analysis of tap water of MNIT

3.4. EXPERIMENTAL PROTOCOL.

Experiments were performed in a batch reactor with a pair of aluminium electrodes Electrodes were connected in a mono polar configuration to an external power supply. The reactor was placed over a magnetic stirrer in each run, to maintain homogeneity inside the batch reactor. When current is passed through the electrodes reactions occurs. The volume of the sample for each batch was 2 litres.

EC experiments were performed until the level of fluoride in the solution is less than the limit suggested by W.H.O for each run and samples was collected after every 10 min from pipette. The treated samples were filtered with Whatman No. 1 filter papers and then analyzed for remaining fluoride concentration. Before each test run electrodes were cleaned with acetone and dilute HCl.

The chemical properties of the prepared sample are listed in Table I. Initial pH of the solution was adjusted to 6 by using HCl for all experiments. It is so because researches have shown that better efficiency is obtained when the pH ranges from 6 – 8. Experiments were carried at room temperature and Fluoride concentration was determined using ion selective electrode method

4. RESULTS

4.1. Effect of current density

Fig. 2. Effect of current density on residual fluoride for an initial fluoride concentration of 4 ppm and at a inter electrode distance of 20 mm ,b) Effect of current density on specific energy consumed for an initial fluoride concentration of 4 ppm and at a inter electrode distance of 20 mm

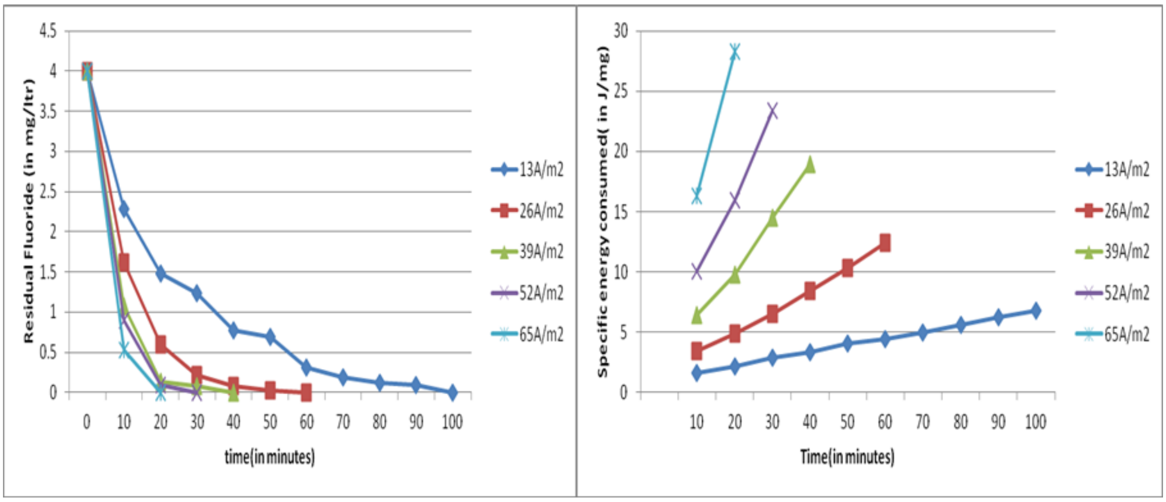
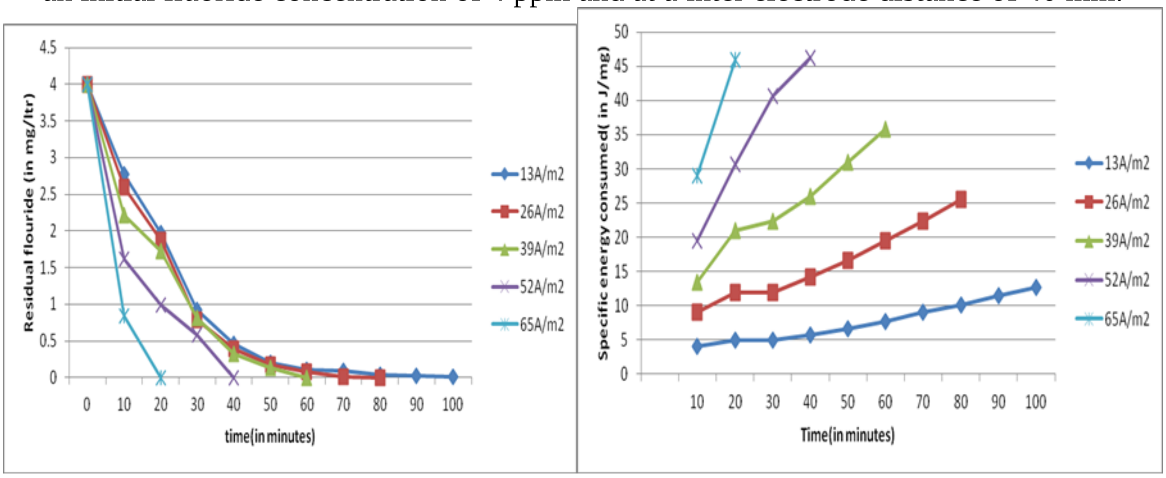
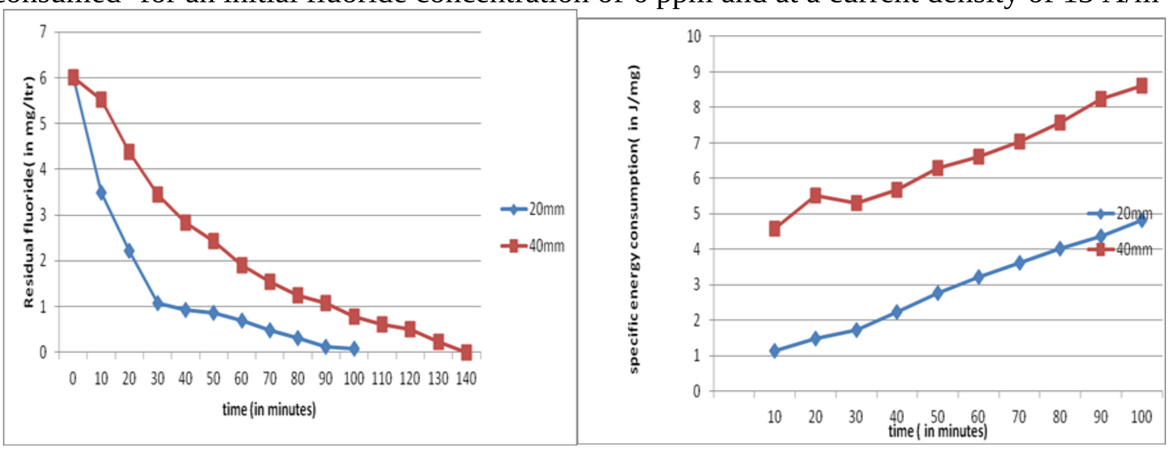


Fig. 3: Effect of current density on residual fluoride for an initial fluoride concentration of 4 ppm and at a inter electrode distance of 40 mm ,b) Effect of current density on specific energy consumed for an initial fluoride concentration of 4 ppm and at a inter electrode distance of 40 mm.



4.2. Effect of Inter electrode distance

Fig. 4 Effect of inter electrode distance on residual fluoride for an current density of 13 A/m^2 and initial fluoride concentration of 6 ppm., b) Effect of inter electrode distance on specific energy consumed for an initial fluoride concentration of 6 ppm and at a current density of 13 A/m^2.



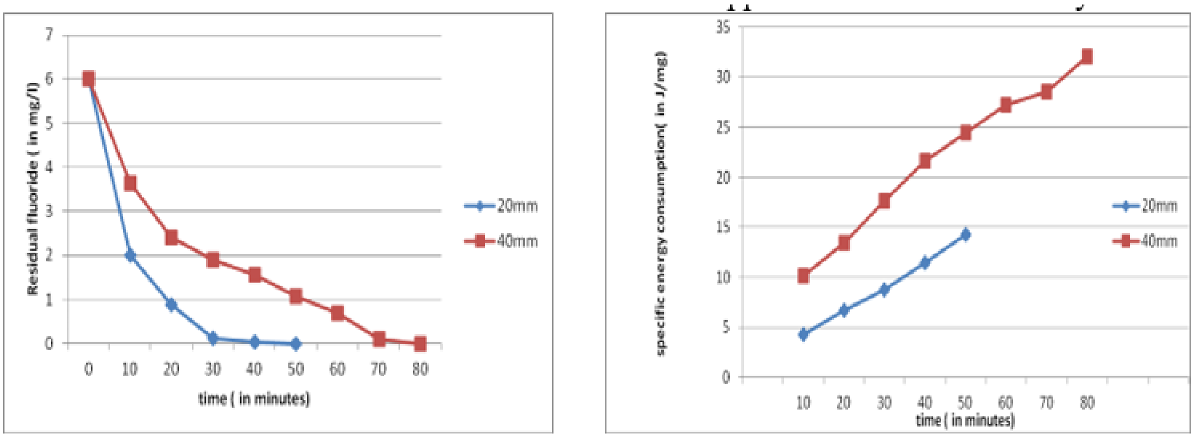


Fig.5. Effect of inter electrode distance on residual fluoride for an current density of 39 A/m^2 and initial fluoride concentration of 6 ppm ,b) Effect of inter electrode distance on specific energy consumed for an initial fluoride concentration of 6 ppm and at a current density of 39 A/m^2.

4.3. Effect of Initial Fluoride Concentration

Fig. 6 Effect of initial fluoride concentration on residual fluoride for a current density of 13 A/m^2 and inter electrode distance of 20 mm ,b) Effect of initial fluoride concentration on specific energy consumed for a a current density of 13 A/m^2 and at a inter electrode distance of 20 mm.

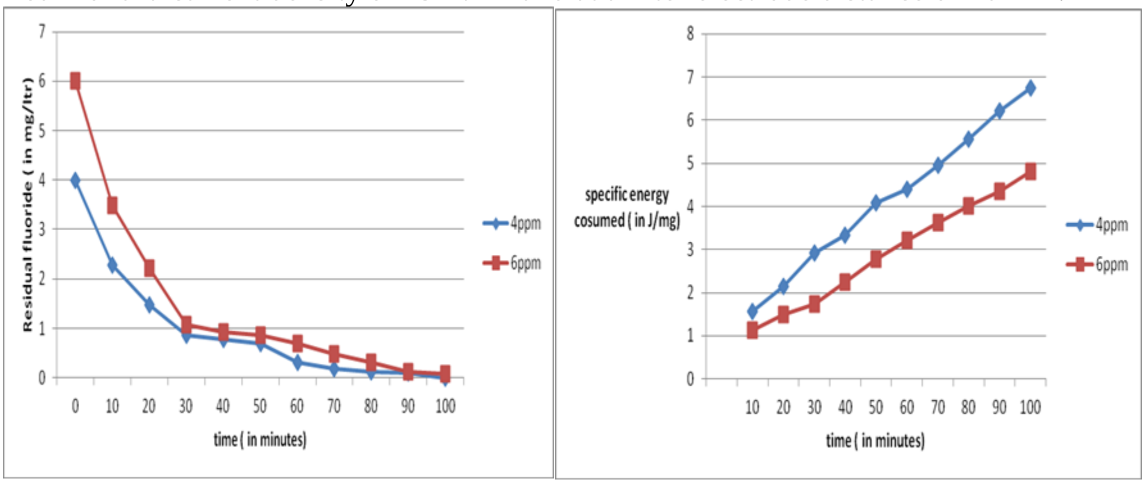
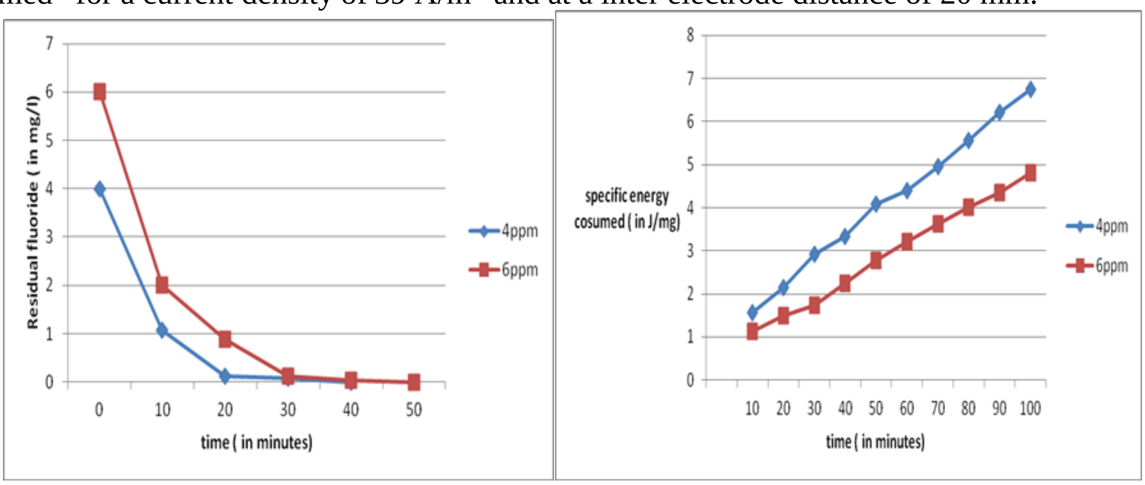


Fig. 7. Effect of initial fluoride concentration on residual fluoride for a current density of 39 A/m^2 and inter electrode distance of 20 mm. b) Effect of initial fluoride concentration on specific energy consumed for a current density of 39 A/m^2 and at a inter electrode distance of 20 mm.



4.4. Experimental Design and Development of Regression Equations

The regression model equation was made between the response and the input variables expressed in terms of coded factors is as follows:

Residual Fluoride = +1.43-0.85*A+0.22* B+0.58* C-0.91* D-0.24 * A * B-0.22 *A*C +0.33* A * D+0.24* B * C-0.081* B * D-0.28 * C * D-0.21 * A * B * C (equation 1)

Specific energy consumed = +26.78 +21.93 * A +5.89* B -5.67 * C +13.07* D +4.09*A * B -6.43*A* C +12.98 * A * D +4.56 * B * D -3.55 *C*D (equation 2)

It is observed that only initial fluoride concentration shows negative effect while current, distance between electrodes and time show positive effect on specific energy consumption.

RUN	Factor1: CURRENT DENSITY, amperes/ m^2 (A)	Factor2: INTER ELECTRODE DISTANCE ,mm(B)	Factor 3: INITIAL FLOURIDE CONCENTRATION, ppm (C)	Factor 4: TIME, min(D)	Response 1: RESIDUAL FLOURIDE, ppm	Response2: ENERGY CONSUMPTION,J/ mg
1	13	40	6	50	2.43	6.3
2	13	40	6	10	5.53	9.57
3	65	40	4	10	0.336	25.07
4	65	20	4	10	0.524	32.58
5	13	20	4	10	2.29	1.57
6	65	20	6	10	1.86	11.59
7	13	40	4	50	0.195	6.6
8	65	40	6	50	0	73.43
9	65	40	6	10	1.89	21.44
10	13	20	4	50	0.685	4.07
11	13	20	6	10	3.48	3.8
12	65	40	4	50	0	114.84
13	13	40	4	10	2.78	4.11
14	13	20	6	50	0.86	2.77
15	65	20	6	50	0	40
16	65	20	4	50	0	70.78

Table 3. Factors and response values of full factorial design

4.4.1. STATSTICLA ANALYSIS

Source	Sum of squares	Df	Mean square	F value	p value Prob > F	
Model	37.54	13	2.89	155.28	0.0064	Significant
A-Current	11.63	1	11.63	625.33	0.0016	
B-Distance between electrodes	0.75	1	0.75	40.28	0.0239	
C-Initial fluoride concentration	5.34	1	5.34	289.96	0.0035	
D-Time	13.18	1	13.18	708.62	0.0014	
AB	0.89	1	0.89	47.97	0.0202	
AC	0.75	1	0.75	40.24	0.0240	

AD	1.76	1	1.76	94.41	0.0104	
BC	0.92	1	0.92	49.51	0.0196	
CD	0.11	1	0.11	5.70	0.1397	
ABC	1.22	1	1.22	65.66	0.0149	
ABD	0.16	1	0.72	38.90	0.0284	
ACD	0.12	1	0.16	8.80	0.0973	
Residual	0.037	2	0.12	6.22	0.1302	
Cor Total	37.57	15	0.019			

Table 4. ANOVA for residual fluoride (mg/l)

Source	Sum of squares	Df	Mean square	F value	p value Prob > F	
Model	16131.05	12	1344.25	40.76	0.0055	Significant
A-Current	7697.43	1	7697.43	233.39	0.0006	
B-Distance between electrodes	554.05	1	554.60	16.82	0.0262	
C-Initial fluoride concentration	514.38	1	514.38	15.60	0.0290	
D-Time	2731.63	1	2731.63	82.82	0.0028	
AB	267.81	1	267.81	8.12	0.0651	
AC	661.78	1	661.78	20.07	0.0207	
AD	2695.69	1	2695.69	81.74	0.0029	
BC	7.51	1	7.51	0.23	0.669	
CD	3321.5	1	332.15	10.07	0.0503	
ABC	202.07	1	202.07	6.13	0.0896	
ABD	374.42	1	374.42	11.35	0.0434	
ACD	91.58	1	91.58	2.78	0.1942	
Residual	98.94	3	32.98			
Cor Total	16229.99	15				

Table 5. ANOVA for specific energy consumed (J/mg)

5. CONCLUSION

1. The study clearly demonstrates that electrocoagulation using aluminum electrodes can be successfully used for removing fluoride from drinking water. A full 2^4 factorial design was undertaken as a design strategy to describe effect of main variables and their interaction on the residual fluoride and specific energy consumption for batch electrocoagulation process. Therefore, effect of current density, initial fluoride concentration, distance between the electrodes and time was evaluated on residual fluoride concentration and specific energy consumption.
2. A mathematical model was developed for residual fluoride and specific energy consumed. From the obtained model it was determined that that current and time show negative effect while initial fluoride concentration and distance between the electrodes show positive effect on the residual fluoride concentration and that current and time show negative effect while initial fluoride concentration and distance between the electrodes show positive effect on the residual fluoride concentration.
3. To check the adequacy of the model ANOVA was adopted where a high R^2 value of 99.90% and 99.39% for residual fluoride and specific energy consumed respectively indicates a good fit between the actual and predicted values. A confirmatory experiment was performed under optimum conditions which showed a close resemblance between the model and experimental results (0.69 mg/l residual fluoride from the experiment compared to 0.77 mg/l from the model). This suggests the use of full factorial design for design of experiment and analysis of data generated thereof.

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