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### Groundwater defluoridation by combined electrochemical coagulation and fungal biosorption process- Part A Groundwater preliminary treatment by batch electrochemical coagulation

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Abstract: Fluoride has beneficial as well as harmful effects on human beings. The combined electrochemical coagulation and fungal biosorption process is an effective treatment option as an alternative to the conventional defluoridation process. In the present study, batch electrochemical coagulation process in bipolar mode using four aluminum electrode was optimized as pre-treatment of combined electrochemical coagulation and biosorption process for defluoridation of groundwater. The batch electrochemical coagulation experiments were carried out by varying operational parameters such as: fluoride initial concentration (4-16mg/L), electrolysis time (10-60min), inter electrode spacing (5-10mm), cell voltage (6-12V) and agitation speed (300-500rpm) to optimize operational parameters for groundwater defluoridation. In addition, the experimental data of batch electrochemical coagulation was analyzed by the Stepwise Multiple Linear Regression analysis (SMLRA) to investigate the most significant operational parameter for the defluoridation of groundwater. The results reveal that the cell voltage and electrolysis time were the most significant operation parameters. The optimum operating parameters for the initial fluoride concentration of 16mg/L are: inter electrode spacing: 5mm, cell voltage: 12V, agitation speed: 500rpm with 91.43% defluoridation efficiency.

**Keywords:** Defluoridation; electrochemical coagulation; Stepwise Multiple Linear Regression Analysis.

#### Introduction

Fluoride contamination in groundwater is of great concern in respect of human health. Fluoride in drinking water has both beneficial and detrimental effects on human health as its concentration within the permissible limit is essential for the bone mineralization and dental enamel formation , whereas above the permissible limit causes multidimensional health manifestations such as dental and skeletal fluorosis<sup>1</sup>. Fluoride contamination in drinking water occurs through natural and anthropogenic sources. Fluoride naturally contaminates groundwater through breakdown of fluoride-bearing rocks (such as fluorospar, fluorite, cryolite, fluorapatite and hydroxylapatite) and soils or weathering and deposition of atmospheric volcanic particles<sup>2</sup>. In addition, wastewater discharges in to water resources from various industries like semiconductor, steel, aluminum, glass, ceramic and fertilizer industries, runoff and infiltration of chemical fertilizers in agricultural

areas contribute to anthropogenic sources of fluoride contamination in groundwater. The permissible limit of fluoride in drinking water, according to the World Health Organization (WHO), Bureau of Indian Standards (BIS) and Indian Council for Medical Research (ICMR) is 0.5- 1.5 mg/L, 1-1.5 mg/L and 1-2 mg/L respectively. To prevent the hazardous effects of fluoride, drinking water fluoride concentrations have to be brought down to < 1.5 mg/L by a quick efficient and safe defluoridation processes.

Till date, many researchers have reported several methods for the removal of fluoride from water i.e., precipitation<sup>3</sup>, ion exchange<sup>4</sup>, adsorption<sup>5</sup>, nanofiltration<sup>6</sup>, electrodialysis<sup>7</sup> and Donnan dialysis<sup>8</sup>. Invariably, most of these methods have disadvantages such as low fluoride removal capacities, high operation and maintenance cost, undesirable effects on water quality, generation of large volumes of sludge and complicated procedures involved in the treatment.

Electrochemical coagulation and fungal biosorption are two promising methods for groundwater defluoridation. Electrochemical coagulation process has advantages over the conventional coagulation process as it is energy efficient, safe, selective, and amenable to automation, cost effective, no chemical requirement and has good fluoride removal efficacy from drinking water even at high fluoride concentration<sup>9</sup>. Electrochemical coagulation process serves as good pre-treatment process for defluoridation. Biosorption is an promising technique for water treatment by using abundantly available biomaterials. Biosorption offers advantages of low operating cost, minimization of the volume of chemical and/or biological sludge , high efficiency in dilute effluents, environmental friendly and economically viable. Therefore, these two methods operated either separately or simultaneously in one –unit results in higher efficiency of fluoride removal. In the combined treatment process, fluoride contaminated groundwater will be first pretreated by electrochemical coagulation process and followed by residual fluoride removal by fungal biosorption.

The present study focuses on optimization of the batch electrochemical coagulation process as pretreatment of combined electrochemical coagulation and biosorption process for defluoridation of groundwater. The significance of operating parameters (viz., initial fluoride concentrations, electrolysis time, inter electrode spacing, cell voltage and agitation speed on defluoridation efficiency was also studied by a Stepwise Multiple Linear Regression Analysis (SMLRA).

#### **Mechanism of Electrochemical Coagulation**

Electrochemical coagulation process is consists of *in situ* coagulant formation as a metallic hydroxides flock within the solution to be cleaned by electrodissolution/electrolytic oxidation of the soluble sacrificial anode material. In ECC process when cell voltage is applied from an external power source, the anode material undergoes oxidation and generates metallic cations, whereas hydrogen gas releases at the cathode, as a result of reduction or reductive deposition of elemental metals. When aluminum electrodes are used, Al<sup>3+</sup> ions generated due to anodic oxidation of electrode, which immediately undergo further spontaneous reactions to form corresponding hydroxides and/or polyhydroxides<sup>10</sup>. These polymeric aluminum hydroxides remove colloids and contaminants by double layer compression of the stern and diffuse layer, charge neutralization, and electroflotation of sweep flocs. The three main reactions that take place in the electrochemical coagulation process while using aluminum electrodes are outlined below<sup>11</sup>:

$Al \rightarrow Al^{3+} + 3e^{-}$ At the anode	(1)
$2H_2O + 2e^- \rightarrow H_2 + 2OH^-$ At the cathode	(2)
$Al^{3+}+3H_2O \rightarrow Al (OH)_3+3H^+$ The hydrolysis reaction	(3)
In addition, there is oxygen evolution (at the anode) reaction:	
$2H_2O + 4e^- \rightarrow O_2 + 4H^+$	(4)

The Al(OH)<sub>3</sub> complexes formed as result of  $Al^{+3}$  hydrolysis (Equation 3) and polymerization, have strong fluoride adsorption capacity and can remove fluoride by forming aluminium fluoride hydroxide complexes (Al<sub>n</sub>F<sub>m</sub>(OH)<sub>3n-m</sub>) by adsorption (Equation 5) and co-precipitation (Equation 6)as shown below:

$Al_n(OH)_{3n} + mF \rightarrow Al_nF_m(OH)_{3n-m} + mOH^-$	Adsorption	(5)
$nAl + (3n - m)OH + mF \rightarrow Al_nF_m(OH)_{3n-m}$	Co-precipitation	(6)

#### **Materials and Methods**

Electrochemical Coagulation is set as a preliminary treatment of combined electrochemical coagulation and fungal biosorption process. Various operating parameters such as cell voltage, initial fluoride concentration,

inter electrode spacing, electrolysis time and agitation speed were studied to optimize the batch ECC for higher defluoridation efficiency. All the batch ECC experiments were carried out with spiked groundwater in bipolar mode using aluminum electrodes.

#### **Characterization of Ground water**

The groundwater used in this study was collected from the campus of Sri Jayachamarajendra College of Engineering, Mysore. The groundwater was characterized for pH, conductivity, chloride, total solids, total dissolve solids, total hardness, iron, fluoride, sulphate and dissolved oxygen by using standard methods<sup>12</sup>. The batch electrochemical coagulation (ECC) was carried out using pre-characterized ground water spiked with the required initial fluoride concentrations of 4, 8 and 16 mg/L. Sodium fluoride (NaF) of Analytical Reagent grade obtained from HiMedia was used to spike water samples to obtain the desired initial fluoride concentration. The physico-chemical characteristics of ground water used to prepare fluoride solution are given in Table 1. The background fluoride concentration in the source water was  $1.04\pm0.02$  mg/L.

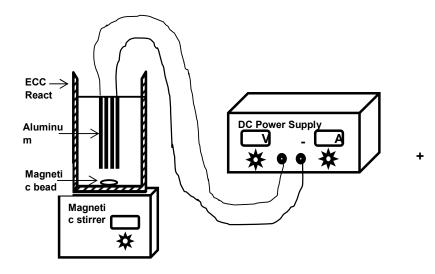
Characteristics Unit	Parameters Value
pH	$6.78 \pm 0.02$
Conductivity (µS/cm)	$1.2 \pm 0.1$
Chlorides (mg/L)	$99.96 \pm 5$
Total Solids (mg/L)	$488 \pm 4$
Total Dissolved Solids (mg/L)	$468 \pm 2$
Total Hardness (mg/L)	$424 \pm 2$
Iron(mg/L)	$0.019 \pm 0.002$
Fluoride (mg/L)	$1.04 \pm 0.02$
Sulphate (mg/L)	37.9 ± 1
Dissolved Oxygen (mg/L)	9 ± 3

#### **Experimental Setup**

The schematic of the laboratory scale batch experimental setup used in the electrochemical coagulation studies is shown in Figure 1. The dimensional characteristics of the experimental setup and the electrical assembly are as shown in Table 2.

Table 2:	Characteristics	of the Electrochemica	l coagulation cell.
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Reactor	
Material	Perspex (organic glass)
Reactor type	batch mode
Shape	Cuboidal
Dimensions (cm)	11.8 x 9 x 17
Agitation mechanism	Magnetic stirrer
Effective volume (L)	1.5
Electrode	-
Material	Aluminum
Shape	Rectangular plate
Size (cm)	9 x 10
Thickness (mm)	1.22
Electrode arrangement	Parallel & bipolar
Effective electrode surface area (cm <sup>2</sup> )	90
Power supply	Direct current (DC)
Voltage range (V)	0-15
Current range (A)	0-6



#### Figure 1: Schematic diagram of ECC experimental setup.

The electrochemical reactor was fed with 1.5 L of groundwater for each experiment. Experiments were carried out using four aluminum electrodes of 9cm x10cm dimension in bipolar mode. The anode and cathode plates were positioned vertically and parallel to each other. Homogenized condition of the reactor contents was maintained throughout the experiments using magnetic stirring unit. A constant inter electrode spacing was maintained throughout the experiment with the help of a non-conducting material. All experiments were carried out at ambient room temperature.

The batch electrochemical coagulation experiments were carried out at varying cell voltage (3-12 V), inter electrode spacing (5 and 10 mm), agitation speed (300 and 500 rpm), electrolysis time (10-60 min) and initial fluoride concentration (4-16 mg/L), each parameter was kept constant for each run. The initial pH of groundwater samples was not varied and experiments were carried out with actual pH values which were in the range of 7.16 to 7.68. The treated samples were drawn at regular intervals of 10 min and filtered using Whatman 42 filter paper for analysing pH and residual fluoride concentration. The fluoride concentration was determined by SPADNS method using Aqualytic fluoride ion analyzer as per APHA 2005.

#### **Results and Discussion**

In the present study, batch ECC process was optimized as preliminary treatment of combined electrochemical coagulation and fungal biosorption process. The effect of each operating parameter on groundwater defluoridation efficiency of batch ECC is discussed in details below:

#### Effect of Cell Voltage

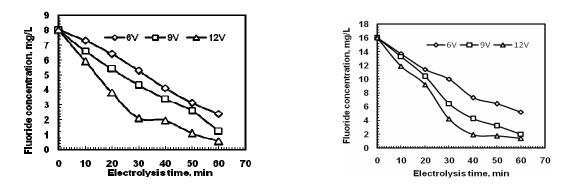


Figure 2: Effect of cell voltage on fluoride removal (Agitation speed = 500 rpm, Inter electrode spacing = 5 mm, Electrolysis time = 60 min, (a)  $F_0$ : 8mg/Land (b)  $F_0$ : 16 mg/L)

Cell voltage as one of the operating parameters; strongly influence the performance of the electrochemical coagulation process. To understand the effect of cell voltage on the groundwater defluoridation efficiency of EC process, cell voltage varied from 6 to 12V by keeping other parameters constant. Figure 2 shows the fluoride degradation curves for  $F_0$  8 and 16 mg/L for applied cell voltages of 6, and 12 V. It was found that, an increase in cell voltage from 6 to 12V lead to an increase in the current density from 0.002A/cm<sup>2</sup> to 0.004A/cm<sup>2</sup>. The defluoridation efficiency increases gradually from 70% ( $F_f$ = 2.4mg/L) to 92.87% ( $F_f$  = 0.57mg/L) and 68.01% ( $F_f$  = 5.2mg/L) to 91.43% ( $F_f$ =1.37mg/L) for the  $F_0$  8 and 16mg/L respectively, with an increase in cell voltage from 6 V to 12 V.

It is clear from the results that defluoridation efficiency increases with the increase in cell voltage. It was also found that at higher cell voltage (i.e., 12V in the present study) fluoride concentration reduces up to the drinking water standard within the electrolysis time of 60min, whereas at the lower cell voltage requires longer electrolysis time (>60min) for the same. It is due to the low current density of  $0.002A/cm^2$  at lower cell voltage (i.e., 6V) to produce sufficient coagulant dose. Similar results were reported by Drouiche *et al.*, 2012 with a finding that applied cell voltage directly determines the coagulant dosage and the rate of bubble generation rates as well as strongly influences solution mixing. The present result also supported by the Ghosh *et al.*, 2008 and Zuo *et al.*, 2008 report that the increase in current density give rise to an increase in charge loading and according to Faraday's law; the amount of aluminum dissolved electrochemically is proportional to charge loadings.

#### **Effect of Initial Fluoride Concentration**

The batch electrochemical coagulation experiments were carried out by varying initial fluoride concentration from 4 to 16 mg/L by keeping other operating parameters constant to determine the effect of initial fluoride concentration on defluoridation efficiency. Figure 3 shows the electrochemical degradation curve for varying initial fluoride concentration  $F_04$ , 8 and 16 mg/L. The fluoride removal efficiency decreases from 97.5% ( $F_f$  = below detection level < 0.1mg/L) to 81.25% ( $F_f$  = 2mg/L) when initial fluoride concentration increased from 4 to 16 mg/L at constant operating condition. Fluoride concentration in the bulk solution influences the extent of electrolysis time, required for achieving the drinking water standards. As seen from the plot, the manifest electrolysis time is shorter for low initial fluoride concentration, while  $F_0$  of 8 and 16 mg/L require higher electrolysis time of 60-75min.

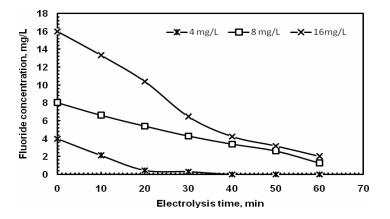
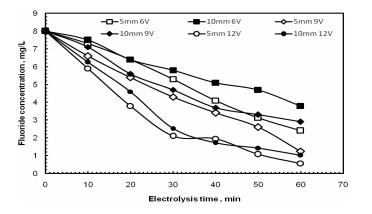


Figure 3: Initial fluoride concentration effect on fluoride removal (Cell voltage=9 V, Agitation speed = 500 rpm, Inter electrode spacing = 5 mm, Electrolysis time = 60 min).

Our results were consistent with the defluoridation result reported by Ghosh *et al.*, 2008; Emamjomeh and Sivakumar 2009 and Essadki *et al.*, 2010. They have reported that in the electrochemical coagulation process, aluminium cations initially contribute to charge neutralization of the pollutant particles as the isoelectric point is attained and lead to lose aggregates formation by sorption coagulation mechanism. As time progresses, further addition of aluminium cation results in amorphous aluminium hydroxide precipitation that promotes pollutant aggregation. At constant current density the production of the aluminium cation remained fixed for varying initial fluoride concentration, therefore at higher initial fluoride concentration, the amorphous aluminium hydroxide was insufficient to form complex with fluoride which lead to increase in final fluoride concentration.



#### **Effect of Inter Electrode Spacing**

## Figure 4:Influence of inter electrode spacing on fluoride removal . (Initial fluoride concentration=8mg/L, Agitation speed = 500 rpm, Electrolysis time = 60 min).

Inter electrode spacing played a significant role in the fluoride removal by electrochemical coagulation. Figure 4 shows electrochemical defluoridation for varying inters electrode spacing of 5 and 10mm. An increase in inter electrode spacing from 5mm to 10mm defluoridation efficiency decreased from 70% ( $F_f$  = 2.4mg/L to 53% ( $F_f$  = 3.8mg/L), 84.37% ( $F_f$  = 1.25mg/L) ) to 77.5% ( $F_f$  = 1.8 mg/L) and 92.87% ( $F_f$  = 0.57 mg/L) to 87.25% ( $F_f$  = 1.02mg/L), at cell voltage 6, 9 and 12 V respectively.

At constant cell voltage, current decreases with the increase in inter electrode spacing due to the decreases in resistance between electrodes<sup>13</sup> as shown in Table 3. At the high current value, the amount of coagulants rises due to the increases in anodic dissolution resulting in higher collisions of the ions that enhance the coagulation. The highest fluoride removal efficiency was achieved with a inter electrode spacing of 5mm by keeping other operating parameter constant.

Electrode spacing, mm	Cell voltage, V	Current, A
	3	0.1
5	6	0.2
	9	0.3
	12	0.4
	3	0
10	6	0.1
	9	0.2
	12	0.3

Table 3: Variation	in current at	t constant :	applied	potential at	t variable inter	electrode spaci	ng
			approx.	porenena a		ereet out space	

#### **Effect of Electrolysis Time**

Electrolysis time is one of the operating parameters that influence fluoride removal efficiency in the electrochemical process. Figure 5 shows the fluoride degradation curves for varying electrolysis time. As seen from the plot, an increase in the electrolysis time from 20 to 90min yields to an increase in fluoride removal efficiency from 21.25% to 77.5 % at applied cell voltage of 6V.

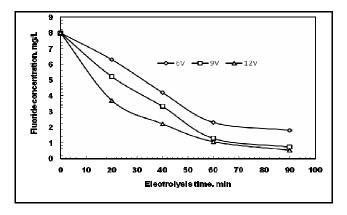
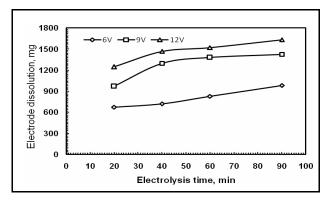


Figure 5: Variation in fluoride concentration with electrolysis time. (Initial fluoride concentration =8 mg/L, agitation speed = 500 rpm, inter electrode spacing = 5mm).



## Figure 6:Effect of electrolysis time on electrode dissolution. (Initial fluoride concentration = 8 mg/L, agitation speed = 500 rpm, inter electrode spacing = 5mm).

Similarly, at applied cell voltage 9 and 12V fluoride removal efficiency increases from 35% to 90.87%, and 53.75% to 93.5% respectively with the increase in electrolysis time from 20 to 90 min. According to Faraday's Law, an increase in the electrolysis time increases the aluminium electrode dissolution hence increases the aluminium cation concentration in solution as seen from Figure 6. Accordingly, with an increase in aluminium cation concentration, the fluoride removal efficiency increased. At longer electrolysis time, amorphous aluminium hydroxide was sufficient to form aluminium fluoride hydroxide complexes  $[Al_nF_m(OH)_{3n-m}]$  as a result, defluoridation efficiency increases.

#### **Effect of Agitation Speed**

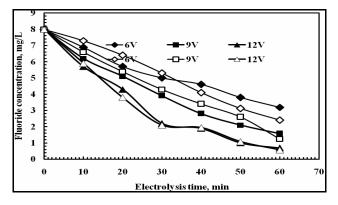
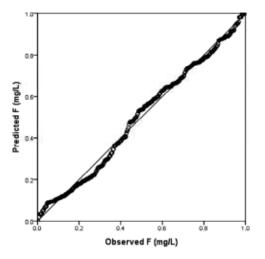


Figure 7: Effect of agitation speed on fluoride removal. (Initial fluoride concentration = 8 mg/L, Inter electrode spacing = 5mm, Electrolysis time = 60min).

The electrochemical coagulation process, also depends on the external mass transfer of coagulants from electrode vicinity to the bulk solution, therefore in the present study effect of agitation speed was evaluated. As seen from Figure 7 at lower cell voltage (6V), defluoridation efficiency was increased by 10% for  $F_0 8mg/L$  with the increase in agitation speed from 300-500rpm whereas at higher cell voltage of 9 and 12V only 4 – 1.5% increase was observed for the same. These results indicate that at higher cell voltage the limiting step of external mass transfer is overcome<sup>16</sup>. Hence in the present study agitation speed was optimized to 500rpm.

#### Effect of pH

The electrochemical coagulation process strongly depends on initial pH of bulk solution. During the electrochemical coagulation of groundwater, pH varies as a result of various electro-chemical reactions involved during the process. In addition, the groundwater typically has a pH value between 6 and 8.5. Fluoride removal efficiency depends on aluminum hydroxide (Al(OH)<sub>3</sub>) floc formation which is an amphoteric hydroxide and pH significantly effects the formation of Al(OH)<sub>3</sub> flocs. Although in the pH range of 5 and 6, Al(OH)<sup>2+</sup> and Al(OH)<sub>2</sub><sup>+</sup> are found as predominant hydrolysis products, Al(OH)<sub>3</sub> is most prevalent in the pH range of 6 and 8.5. Figure 8 shows changes in pH of the groundwater having different fluoride concentration during electrolysis.



# Figure 8: Groundwater pH variation with respect of electrolysis time during the batch electrochemical coagulation process(Cell voltage=9 V, Agitation speed = 500 rpm, Inter electrode spacing = 5 mm, Electrolysis time = 60 min).

At the end of 60 min electrolysis time, pH reduction by at least one unit was observed for each fluoride concentration. Although, pH reduction was observed, the final pH values after treatment were found within the BIS standard of 6.5-8.5. The reason for a relatively low pH is ascribed to the  $H^+$  ion (generation near the anode) that remain in the solution after electrochemical coagulation, according to the equation:

$$Al^+ + 3H_2O \rightarrow Al (OH)_3 + 3H$$

(7)

The dried floc materials tend to have a pH value in the alkaline range. The formation of  $Al(OH)_3$  as the electrolysis progress (pH 5-7), lowers the solution pH as it reduces the moles of hydroxyl ions in the solution. In addition to this effect, oxygen gas evolution at the anode, because of its corrosively results in the decrease in final pH value. In the alkaline condition, pH decrease is also ascribed to the formation of hydroxide precipitate with other cations; the formation of  $Al(OH)_4^-$  adds to the buffering capacity.

In addition, the presence of chloride ions (or TDS) in groundwater also influences the decrease in final pH after electrochemical coagulation because chloride oxidation also occur in a parallel reaction along with anodic dissolution of aluminum, Chlorine gas evolved on the surface of the anode (Equation 8) and the reactions occurring in the bulk solution (Equation 9 and 10)<sup>17</sup>. Once dissolved, chlorine disproportionate to produce hypochlorous and hydrochloric acids in the solution. The acids dissociate and neutralize the OH<sup>-</sup> generated on the cathode, thereby lowering the solution pH.

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$Cl_2+H_2O \rightarrow HOCl + H^++Cl^-$	(9)
$HOCI \rightarrow H^+ + OCI^-$	(10)

#### Stepwise Multiple Linear Regression Analysis (SMLRA) of data

Stepwise Multiple Linear Regression Analysis of batch ECC data was performed in order to examine the effect of each predictor variable (i.e., initial fluoride concentration ( $F_0$ ), electrolysis time (ET), inter electrode spacing (IES), cell voltage (CV) and agitation speed (AS) on the ground water defluoridation efficiency. SMLRA sequentially enters the predictor variables and retains or removes them based on their constant value assessment, which ends up with a model having the least possible set of predictor variables. SMLRA exclude the subsets of predictor variables like "agitation speed" as shown in Table 4, because they were statistically insignificant (p>0.05). It infers that "agitation speed" does not play a significant role in defluoridation of ground water by ECC treatment. The predicted values and the observed values of criterion variable showed highest correlation of 0.847 in 'model 4' with 71.7% R Square value, 71.3% of adjusted R Square value and a least standard error of 2.12080 as shown in Table 5.

	Model	Beta In	Т	Sig.
1	F <sub>0</sub> CV AS IES	0.538 0.005 - 0.009 0.130	16.135 0.110 - 0.204 2.934	0.000 0.913 0.838 0.004
2	CV AS IES	- 0.261 - 0.009 0.130	-7.757 - 0.274 3.985	$0.000 \\ 0.784 \\ 0.000$
3	AS IES	- 0.009 0.130	- 0.299 4.358	0.765 0.000
4	AS	- 0.009	- 0.307	0.759

Table 4: Excluded Variables from model of Stepwise Multiple Linear Regression Analysis (SMLRA).

Table 5: Summary of Models generated by Stepwise Multiple Linear Regression Analysis (SML	RA) of
batch electrochemical coagulation data.	

Model	R	R Square	<b>Adjusted R Square</b>	Standard Error of the
				Estimate
1	0.595 <sup>a</sup>	0.354	0.352	3.18716
2	0.802 <sup>b</sup>	0.643	0.641	2.37201
3	0.837 <sup>c</sup>	0.700	0.697	2.17960
4	$0.847^{d}$	0.717	0.713	2.12080

a. Predictors: (Constant), ET, b. Predictors: (Constant), ET,  $F_0$ , c. Predictors: (Constant), ET,  $F_0$ , CV, d. Predictors: (Constant), ET,  $F_0$ , CV, IES.

It is evident that 'model 4' explained 71.3% proportion of variation in ECC process by four predictors ( $F_0$ , ET, IES and CV) out of the other three respective models. The Regression Coefficients are the "Standardized Beta Coefficients" which illustrate the relationship between the standardized dependent variable and the standardized independent variable. Beta coefficients elucidate the respective importance of each independent variable in the Multiple Linear Regression Model. The negative Beta coefficient (-0.635) of "electrolysis time" and "cell voltage" (-0.261) ascertained that their higher values are related to the lower values of final fluoride concentration (FFC) as shown in Table 6.

	Model	Std. Error	Standardized Coefficients Beta	t	Sig.
4	(Constant)	0.537	4.967	9.244	0.000
	ET	0.004	- 0.635	-21.233	0.000
	F <sub>0</sub>	0.025	0.646	19.728	0.000
	CV	0.046	- 0.261	-7.973	0.000
	IES	0.047	0.130	4.358	0.000

Table 6: Standardized Beta Co-efficient of Model 4

The positive Beta coefficient (0.130) of "inter electrode spacing" and (0.646) of "fluoride initial concentration" established a relationship that their higher values are associated with the higher values of final fluoride concentration. The best suited MLR model is given below:

#### FFC (mg/L) = 4.967 - 0.635 (ET) + 0.646 (F<sub>0</sub>) - 0.261 (CV) + 0.130 (IES) (11)

The predicted final fluoride concentration values and the observed final fluoride concentration values after ECC process were plotted in Figure 9. The T value of regression coefficients of dependent variables determined that "electrolysis time" and "cell voltage" were seen to be the most significant parameters of the ECC process for defluoridation of ground water. The P value (P<0.05) of electrolysis time, initial fluoride concentration, cell voltage and inter electrode spacing were found to be statistically significant at the 5% probability level on defluoridation of ground water by electrochemical coagulation.

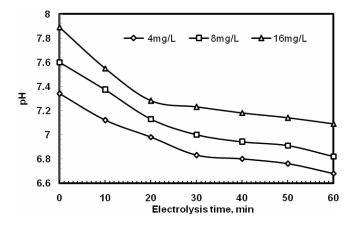


Figure 9: Predicted and observed final fluoride concentration (mg/L).

#### Conclusion

Batch electrochemical coagulation (ECC) was investigated for the removal of excess fluoride from groundwater using bipolar electrode connections as pretreatment of combined electrochemical coagulation and biosorption process. Effects of cell voltage, initial fluoride concentration, inter electrode spacing, electrolysis time and agitation speed on the defluoridation efficiency was studied in detail for the optimization of operating parameters. The optimized parameters for batch ECC was initial fluoride concentration of 16mg/L, inter electrode spacing; 5mm, cell voltage; 12V, agitation speed; 500rpm and natural pH of ground water with 83.7%defluoridation efficiency. The results showed that the defluoridation efficiency of ECC process depends strongly on cell voltage and electrolysis time as removal efficiency increases with the increase in cell voltage and electrolysis time. But there is a negative impact of inter electrode spacing and initial fluoride concentration lesser the fluoride removal efficiency.

Stepwise Multiple Linear Regression Analysis was performed to choose the significant predictor variables and rank them in accordance to their influence in ground water defluoridation by electrochemical coagulation process. The analysis excluded "agitation Speed" for being insignificant, inferring poor impact on the ECC process. "Model No. 4" could explain 71.3% of variance in final fluoride concentration by denoting

"Electrolysis Time" and "Cell Voltage" as the chief predictors influencing the defluoridation efficiency, where as "Inter Electrode Spacing" and "Initial Fluoride Concentration" are the subordinate predictors with minor impact on the same process.

## Furthermore batch ECC treated groundwater was subjected to post treatment by fungal biosorption for residual fluoride removal.

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