

Electrochemicalcoagulation Treatment in Simultaneous Arsenite and Fluoride Removal from Drinking Water

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ABSTRACT: This research paper focus on the role of placing position ofelectrodes inside the electrochemical coagulation reactor (ECCR) for simultaneous removal of arsenite and fluoride from drinking water. Experiments for different electrode arrangement such as Fe₁-Al₂-Al₃-Fe₄;; Al₁-Fe₂-Fe₃-Al₄, Al₁-Fe₂-Fe₃-Fe₄ and Fe₁-Fe₂-Fe₃-Al₄ were carried out for pre-optimized operating condition in batch electrochemical coagulation (BECC) studies using 4 electrodes giving a specific area to volume (SA/V) ratio of 40 m²/m³ treated with cell voltage of 16V for 45 min electrolysis time (ET). The initial fluoride and arsenite concentration in groundwater were 12 mg/L and 1.6 mg/L respectively. The maximum simultaneous As(III) and F⁻ removal was achieved for Fe₁-Al₂-Al₃-Fe₄ electrode combination still by retaining the water quality parameters within the drinking water standards proving the effectiveness of the electrochemical coagulation as a novel option for simultaneous removal of arsenite and fluoride from contaminated waters using a single technology.

KEYWORDS: Arsenite, Fluoride, Aluminum and Iron electrodes, Electrochemical coagulation treatment.

I. INTRODUCTION

Safe water is a fundamental human right and it's a basic need of each individual. Arsenic-contaminated water is a significant problem owing to its toxicity. In India, most villages and towns use groundwater as drinking water source. Currently, government interventions to solve arsenic and fluoride contaminated groundwaters are deep dreams though Environmental (Protection) Act of 1986 is observed as a partial mandate limited to industries. Long-term exposure to arsenic leads to chronic health problems such as hyperpigmentation and keratosis of the hands and feet; it also causes bladder, lung, skin, kidney, liver, and prostate cancer [1]. Considering the high toxicity of arsenic, World Health Organization (WHO) and the US Environmental Protection Agency have set a maximum acceptable level of 10 µg L⁻¹ for arsenic in drinking water. An excess amount of fluoride anions in drinking water is known to cause devastative negative effects on human health. To prevent these harmful consequences, especially problems resulting from fluorosis, the World Health Organization (WHO) fixed the maximum acceptable value of fluoride anions in drinking water limited to 1.5 mg L⁻¹ [2].

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Different technologies have been developed to decrease the arsenic and fluoride concentrations in groundwater, such as reverse osmosis, electrodialysis and distillation [3,4]. For arsenic removal, adsorption [5] and coagulation-flocculation procedures are most common involving the addition of aluminum sulfate, ferric sulfate, calcium hydroxide or polymeric flocculants. However, due to low concentrations of dissolved matter these processes give off large quantities of sludge, because SO_4^{2-} anions consume 50% of the coagulant [6]. Moreover, it is well known that arsenite oxidizes to arsenate on the anode during the electro-chemical coagulation process [7].

The most widespread method for removing excess fluoride from groundwater is the coagulation-flocculation process using calcium and aluminum salt. Nevertheless, this process causes an increase in pH, soluble aluminum species which are toxic to human health. Besides when treating large volumes of water this process yields large amounts of sludge resulting in waste management problems[8].

II. LITERATURE SURVEY

Electrochemical coagulation(ECC) was chosen as to provide one novel technology to remove both arsenic and fluoride from contaminated waters. In the ECC process, aluminum or iron is dissolved electrolytically and the electrolytically generated flocs of metallic hydroxides remove the dissolved species [9-11]. The electrode materials commonly used for the removal of Arsenic by EC are mainly iron [12] and aluminum [13]. [14] demonstrated that arsenic removal from drinking water was better using aluminum electrodes than iron electrodes.

ECC known as an effective treatment process destabilize dispersed fine particles and anions in water [15-16]. Electro-dissolution of aluminum generates bulk aluminum hydroxides ($\text{Al}(\text{OH})_{3(s)}$) and aluminum oxides ($\text{Al}_2\text{O}_{3(s)}$), which adsorb arsenic and remove fluoride by substitution reactions on the surface of the aluminum hydroxides [17,18].

Few studies have shown simultaneous removal of arsenic and fluoride [19], where iron and aluminum binary oxide (FeAlO_xH_y) [20,21] and freshly-prepared aluminum hydroxide are employed as adsorbent materials. It is important to that the main interference observed during simultaneous removal of arsenic and fluoride using freshly-prepared aluminum hydroxide is the formation of soluble complexes of Al-Fe, which solubilize aluminum oxyhydroxides (AlO_xH_y) increasing the concentration of residual aluminum. [20] reports the efficient removal of both arsenic and fluoride simultaneously from synthetic solutions using a combination of both electro-oxidation and EC processes.

The electrochemical system has been applied to many water or wastewater treatment processes. We propose one technology for simultaneous treatment of arsenic and fluoride contaminated water. Therefore, the objective of the present research study was to evaluate the role of placing position of electrodes inside the ECR using electrode combinations for simultaneous removal of fluoride and arsenite from drinking water.

III. MATERIALS AND METHODOLOY

Chemicals: All the chemicals used in the study were of analytical reagent (AR) grade. Sodium meta arsenite, sodium fluoride, zirconyl chloride octahydrate, SPADNS were procured from Hi-Media Laboratories Pvt. Ltd., Mumbai. Silver nitrate, potassium chromate, ammonium chloride, sodium hydroxide, ferrous ammonium sulphate, ammonium acetate, hydroxylamine HCl, sodium chloride was procured from S.D. Fine-Chemicals limited, Mumbai. White phenol, sodium thiosulphate, ammonium molybdate and ammonium vanadate were purchased from Qualigens Fine Chemicals, Mumbai. Potassium iodide crystals, starch indicator, methyl orange indicator and sulphuric acid were obtained from Merck specialities, Mumbai.

Analytical Methods: Physico-chemical water quality parameters were analyzed in accordance with the Standard Methods (APHA et al. 2005). pH was measured using a pH meter (LI 127 Elico). Electrical conductivity of the solution was measured using a digital conductivity meter (Systronics Model 30/10FT YSI). Temperature variations in the bulk solution was recorded using digital thermometer. Iron, nitrate, fluoride, sulphate and phosphate concentrations in the water was measured using a spectrophotometer. Total hardness, total alkalinity as CaCO_3 and total dissolved solids (TDS) were determined following procedures described in the Standard Methods (ALPHA). Arsenite in the form of arsenic and residual aluminum ion concentration in the solution was determined using Inductively Coupled Plasma (ICP Horiba JobinYvon, France).

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Experimental Setup: The laboratory scale batch electrochemical coagulation(ECC) reactor unit was designed and fabricated using a see-through material (acrylic glass) giving an effective working volume of 2 Liters. The ECC reactor consists of electrode holding arrangements to keep electrode plates parallel to each other face to face between the walls of the ECR to maintain equal anode-cathode face or maintain a perfect area. The electrodes were washed with 15% HCl solution before conducting each experiment to remove any dirt. These electrodes were placed parallel to each other with a predetermined inter-electrode spacing of 5mm in a bipolar connection. The current input to the ECR and the corresponding cell voltage was maintained constant by means of a precision DC power supply (Textronix 35D, Dual regulated power supply, 0-16V, 0-10A) unit. The DC power supply unit was operated for applied cell voltages 16V for 4 electrode numbers for 45 min of electrolysis time which is the optimized operating condition obtained through a series of experiments not reported here. Uniform mixing of the bulk content of the ECR was achieved by all inductive magnetic stirrer (REMI 2MLH). All ECC test runs were performed at ambient room temperature. In each discrete run, 2L of arsenite and fluoride spiked groundwater of known initial concentration $As(III)_0$ 1.6 mg/L and F_0^- 12 mg/L was fed into the ECC reactor. Experiments were carried out for the 16V, 4E for 45 min with intent to observe the role of electrode placing position in ECR for simultaneous removal of both arsenite and fluoride.

Samples were retrieved at regular time intervals, filtered and analyzed for residual arsenite, iron and aluminum concentration using inductively coupled plasma (ICP) and fluoride concentration were determined using spectrophotometer after each ECC run for a pre-determined electrolysis time (ET). At the end of each experimental run, the electrodes were washed thoroughly with water followed by 15% HCl solution and again rinsed in distilled water to remove trapped floc/ solid residues in the electrode valleys and dents on its surfaces, then dried and weighed. The difference in weight provides information on the anode dissolution useful in determining the operating cost.

V. EXPERIMENTAL RESULTS

Whiling using two different electrode materials its compatibility and combined effect on treatment influences the performance of the ECC process. Appropriate selection of the electrode material and its position in the ECR play an vital role in delivering proper M^+ ions in such a way that it facilitates simultaneous removal of fluoride and arsenite from water. Four different electrode combinations such as (i) $Al_1-Fe_2-Fe_3-Al_4$ (ii) $Al_1-Fe_2-Fe_3-Fe_4$ (iii) $Fe_1-Fe_2-Fe_3-Al_4$ and (iv) $Fe_1-Al_2-Al_3-Fe_4$ are explored. Batch ECC studies were carried out for the pre-optimised operating condition using 4 Nos. of electrodes giving SA/V ratio of $40 m^2/m^3$ with fixed inter-electrode spacing of 5 mm at applied cell voltage of 16V for 45 min ET.

4.1 Effect of $Fe_1-Al_2-Al_3-Fe_4$ arrangement electrodes for simultaneous removal of arsenite and fluoride

Fig. 1 shows the variation in concentration of fluoride, arsenite, residual aluminum and iron during the electrolysis process as a function of ET.

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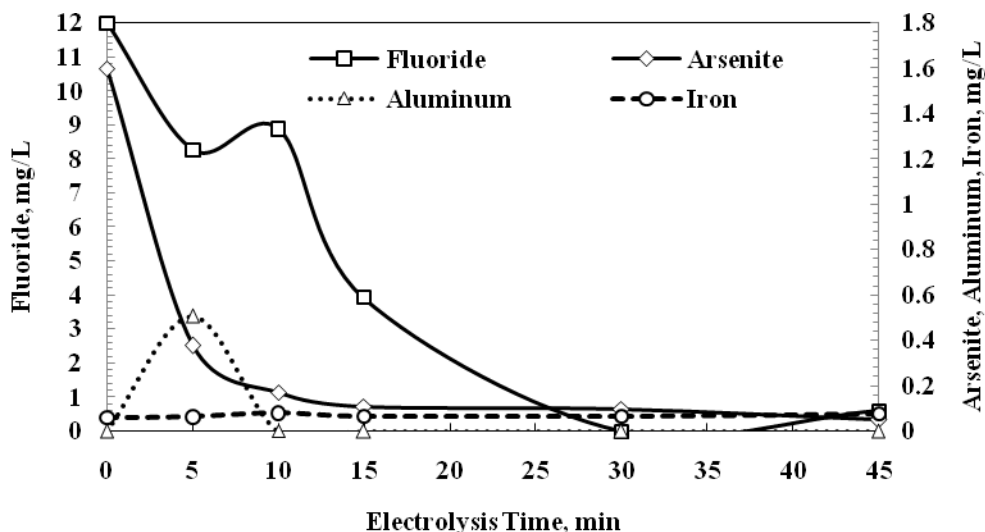


Fig: 1 Fluoride and arsenite removal using Fe₁-Al₂-Al₃-Fe₄

Operating Conditions: Bipolar Fe₁-Al₂-Al₃-Fe₄, E. no: 2Fe+2Al, pH₀: 7.99, As(III)₀: 1.6 mg/L, F₀⁻: 12 mg/L, Al₀³⁺: 0mg/L in solution, Fe₀: 0.061 mg/L in solution, E spacing: 5mm, cell voltage: 16V, average current: 0.67A, SA/V ratio: 40 m²/m³ and ET:45 min.

Iron electrode is used as feeder anode and cathode in the first position linked to the positive terminal of DC unit and by placing aluminum electrodes in between them. In this arrangement after 5 min ET, the fluoride concentration reduced from 12 mg/L to 8.28 mg/L and arsenite reduced from 1.6 mg/L to 0.378 mg/L with simultaneous increase in bulk solution pH from 7.53 to 7.64. Changes in color of the bulk solution from colorless to pale yellow indicate dissolution of iron and aluminum electrodes during electrolysis. The color imparted by Fe²⁺ and Fe(OH)₂ with pale/green. From 5-10 min ET, fluoride removal declined because of insufficient Al(OH)₃ floc formation to trap the F⁻ ions and desorption. Arsenite concentration was reduced from 0.378 to 0.172 mg/L indicating higher efficiency of using iron electrodes as anode. After 30 min ET fluoride values reduced to 0 mg/L and arsenite concentration remained constant from 10 - 45 min with bulk solution color turbid white with light green tinge showing pH value of 7.4 indicating the Al(OH)₃ formation was more dominant over the Fe(OH)₂ concentration during this period. In the bulk solution as a evidence the residual Al³⁺ ion concentration was found to be 0 mg/L indicating the occurrence of a complete sweep coagulation process with adsorption of F⁻ ions from groundwater, however As(III) concentration of 0.1 mg/L remained constant upto 45 min ET. F⁻ and As(III) concentration at 45 min ET was 0.61 and 0.05 mg/L, As(III) almost within permissible limit prescribed by BIS drinking water quality standards. This prompted us to replace aluminum as anode and cathode by placing two iron electrodes in between them.

4.2 Al₁-Fe₂-Fe₃-Al₄ arrangement electrodes for simultaneous removal of arsenite and fluoride

This order of electrode arrangement indicates that Al 2D plates are positioned in the extremes and two Fe electrode in between them in the ECR. Fig. 2 shows variation in concentration of fluoride, arsenite, residual aluminum and iron during the electrolysis process as a function of ET. During the first 5 min of electrolysis the F⁻ and As(III) concentration reduced from 12 mg/L to 10.1 mg/L and from 1.6 mg/L to 0.19 mg/L with decrease in pH from 7.54 to 7.44 because of acidic conditions at the anode face by the effect of H⁺ ion released with the solution color turning pale yellow indicating Fe³⁺ formation. After 5 min ET the solution color changed to green due to formation of Fe(OH)₂ flocs resulting in F⁻ and As(III) reductions to 4.14 mg/L and 0.019 mg/L respectively. The pH of the bulk solution decreased from 7.54 to 7.24 with the increase in temperature from 25°C to 27°C because of multiple exothermic reactions in the cell leading to decrease in conductivity values.

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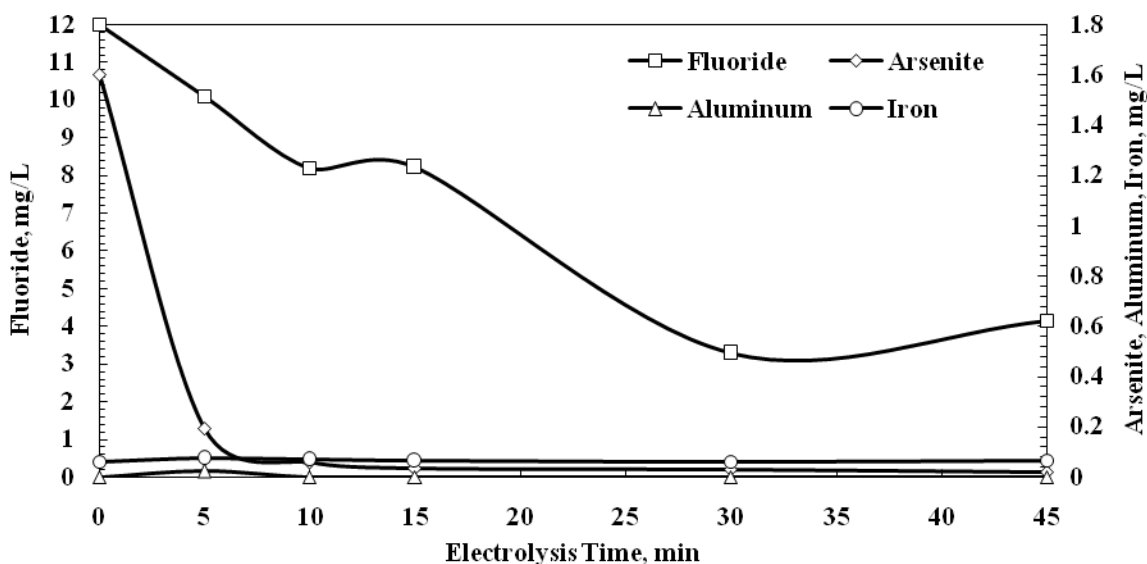


Fig. 2 Fluoride and arsenite removal using Al₁-Fe₂-Fe₃-Al₄

Operating Conditions: Bipolar Al₁-Fe₂-Fe₃-Al₄, E. no: 2Fe+2Al, pH_0 : 7.99, $As(III)_0$: 1.6 mg/L, F_0^- : 12 mg/L, Al_0^{3+} : 0 mg/L in solution, Fe_0 : 0.061 mg/L in solution, E spacing: 5mm, cell voltage: 16V, average current: 0.67A, SA/V ratio: 40 m²/m³ and ET :45 min.

The residual aluminum and iron concentration in the treated water after 45 min ET were 0 and 0.066 mg/L much less than the limits prescribed by the Bureau of Indian Standards (BIS). Thus, Al₁-Fe₂-Fe₃-Al₄ electrode arrangements in simultaneous removal of fluoride and arsenite could not be achieved in 45 min of ET.

4.3 Effect of using Al₁-Fe₂-Fe₃-Fe₄ arrangement electrodes for simultaneous removal of arsenite and fluoride

Fig. 3 shows changes in fluoride, arsenite, aluminum and iron concentration during electrolysis. Fluoride concentration reduced from 12 mg/L to 2.6 mg/L, arsenite from 1.6 mg/L to 0.027 mg/L with simultaneous marginal decrease in pH from 7.57 to 7.54. The bulk solution color changed from colorless to turbid white at 45min ET following the reaction shown in equation (1).



After 10 min ET the solution turned to pale green color as a result of equation (2).



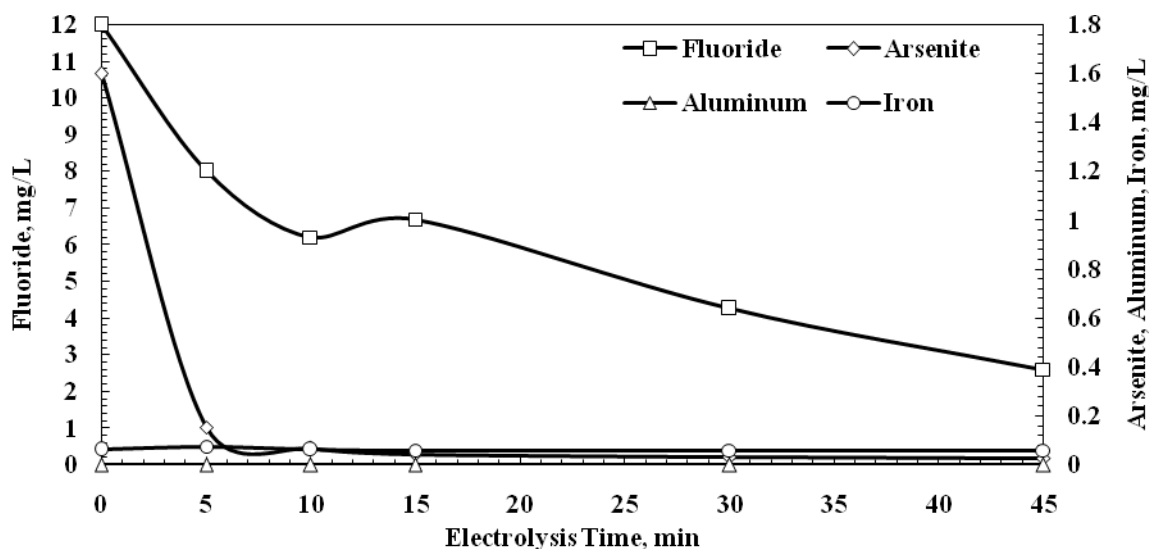
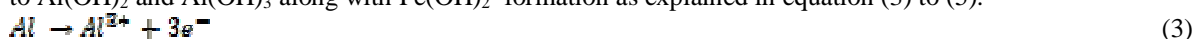


Fig. 3 Fluoride and arsenite removal using Al₁-Fe₂-Fe₃-Fe₄

Operating Conditions: Bipolar Al₁-Fe₂-Fe₃-Fe₄, E. no: 1Al+3Fe, pH₀: 7.99, As(III)₀: 1.6 mg/L, F₀⁻: 12 mg/L, Al₀³⁺: 0 mg/L in solution, Fe₀: 0.061 mg/L in solution, E spacing: 5mm, cell voltage: 16V, average current: 0.67A, SA/V ratio: 40 m²/m³ and ET:45 min.

From, equation(2) the H⁺ ions liberate into the solution resulting in pH decrease along with formation of Fe(OH)₂ imparting green color. At 15 min ET the pH was 7.62 with turbid white and a green tinge. The turbid white color is due to Al(OH)₂ and Al(OH)₃ along with Fe(OH)₂ formation as explained in equation (3) to (5).



At 30 min ET the solution appeared turbid white with the formation of precipitate because of aluminum ions in suspension. Residual aluminum and iron concentration after treatment in the treated water was 0.01 mg/L and 0.061 mg/L respectively. Thus, Al₁-Fe₂-Fe₃-Fe₄ electrode arrangements for simultaneous removal of fluoride and arsenite could not be in 45 min ET thereby ET needs to be extended.

4.4 Effect of using Fe₁-Fe₂-Fe₃-Al₄ arrangement electrodes for simultaneous removal of arsenite and fluoride

Electrode placing positions were changed as shown in Fig.4. Fluoride concentration reduced from 12 mg/L to 6.9 mg/L and arsenite from 1.6 mg/L to 0.005 mg/L, residual aluminum and iron concentration was found to be 0.041 and 0.059 mg/L at 45 min ET. pH decreased from 7.74 to 7.61 with change in bulk solution color from colorless to pale yellow- green, turbid white and finally colorless.

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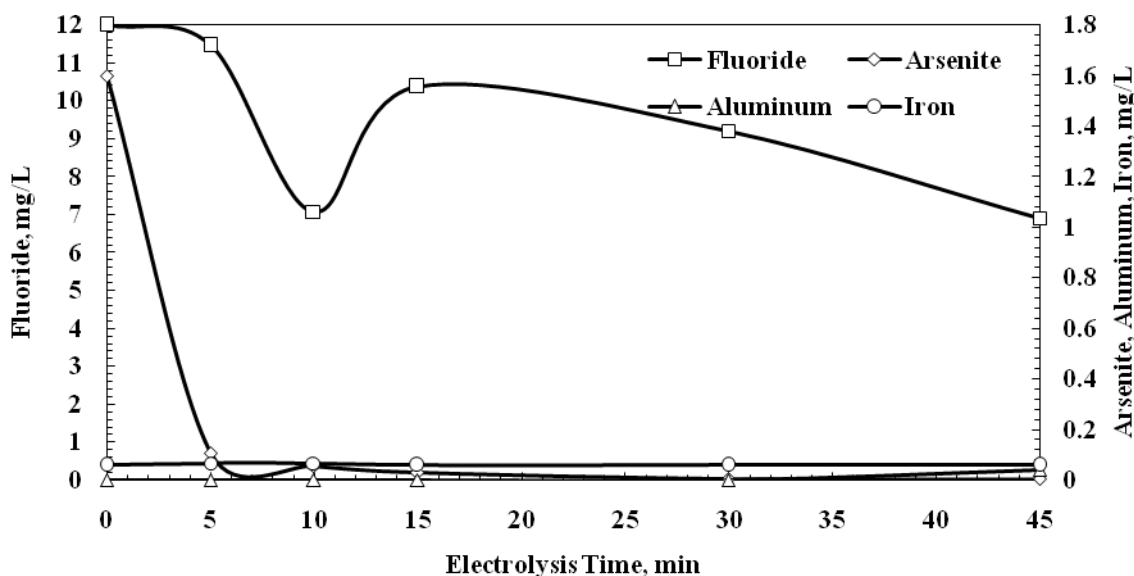


Fig.4 Fluoride and arsenite removal using Fe₁-Fe₂-Fe₃-Al₄

Operating Conditions: Bipolar Fe₁-Fe₂-Fe₃-Al₄, E. no: 3Fe+1Al, pH₀: 7.99, As(III)₀: 1.6 mg/L, F₀⁻: 12 mg/L, Al₀³⁺: 0 mg/L in solution, Fe₀: 0.061 mg/L in solution, E spacing: 5mm, cell voltage: 16V, average current: 0.67A, SA/V ratio: 40 m²/m³ and ET:45 min.

The colorless solution of the bulk solution is because of the formation of hypochlorous acid (HOCl) in the ECC reactor as a result of complex chemical coagulation reactions that remove contaminants and colloidal particles from water. At slightly acidic pH, the chloride ions present in the solution as HOCl have higher oxidation potential (1.49V) so results in decolorization of the solution[23]. With the above observations, it was concluded that Fe₁-Fe₂-Fe₃-Al₄ was unable to remove arsenite and fluoride from water simultaneously within 45 min of ET requiring longer ET to achieve well within the drinking water standards.

4.5 Post ECC drinking water quality parameters

ECC being a novel treatment technology for removal of target contaminants still retaining the beneficiary mineral content of water. Drinking water quality parameters before and after ECC treatment of arsenite and fluoride contaminated water ECC was filtered using whatman filter paper and subject to physico-chemical parameters analyses following Standard Methods. The results of the analyses is shown in Table 1.

Table 1 Water quality constituents before and after the ECC treatment process

Sl. No.	Water Quality Parameter	Unit	BIS-10500 - 2001	Water Quality parameter value before ECC	After 45 min of ECC for operating conditions: 16V; As(III) ₀ : 1.6 mg/L; F ₀ ⁻ : 12 mg/L			
					Fe-Al electrode combination			
					Fe ₁ -Al ₂ -Al ₃ -Fe ₄	Al ₁ -Fe ₂ -Fe ₃ -Al ₄	Al ₁ -Fe ₂ -Fe ₃ -Fe ₄	Fe ₁ -Fe ₂ -Fe ₃ -Al ₄
1	pH	-	6.5-8.5	7.52-7.73	7.4	7.24	7.54	7.61
2	Total Alkalinity as CaCO ₃	mg/L	300 - 600	360-377	204	220	200	204

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3	Chloride	mg/L	250-1000	70-91	80.23	76.32	72.40	74.36
4	Total Hardness as $CaCO_3$	mg/L	300-600	352-463	168	192	216	184
5	Iron	mg/L	0.3-1.0	0.01-0.03	0.078	0.066	0.061	0.059
6	Aluminum	mg/L	0.03 – 0.2	0.0	0.0	0.0	0.0	0.04
7	Fluoride	mg/L	1 - 1.5	12	0.61	4.14	2.6	6.9
8	Arsenic	mg/L	0.01-0.05	1.6	0.05	0.019	0.027	0.005

From the table, it may be observed that the pH value after ECC were well under control because a combination of Fe and Al electrodes were used. Total hardness values were removed by over 50%, Over 50% of total alkalinity were removed after ECC within safe limits set by BIS. Chloride values were more or less the same before and after ECC. Iron concentration in treated water were much below the lower limits of BIS standards, while aluminum concentrations were zero mg/L. Except arsenite and fluoride all other water quality parameters after ECC was well within the BIS drinking water standards.

V. CONCLUSION

A single solo ECC treatment technology has proved is novelty in removing two dangerous contaminants arsenite and fluoride from drinking water still retaining other beneficial quality parameters intact. The role of interchanging the electrode placing position in ECR in simultaneous removal of As(III) and fluoride removal were investigated. The study was carried out for $Fe_1-Al_2-Al_3-Fe_4$; $Al_1-Fe_2-Fe_3-Al_4$; $Al_1-Fe_2-Fe_3-Fe_4$ and $Fe_1-Fe_2-Fe_3-Al_4$ electrode arrangement combination for predetermined operating condition with applied cell voltage of 16V, 4 Nos. of electrodes for 45 min of ET with initial As(III) and concentration of 1.6 mg/L and 12 mg/L respectively. The study revealed that and As(III) was reduced to 0.61 mg/L and 0.005 mg/L for $Fe_1-Al_2-Al_3-Fe_4$ and $Fe_1-Fe_2-Fe_3-Al_4$ combination individually. The maximum simultaneous removal of both As(III) and of 96.87% and 94.91% for $Fe_1-Al_2-Al_3-Fe_4$ combination could be achieved within 45 min of ET.

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