



## Factors influencing arsenic and nitrate removal from drinking water in a continuous flow electrocoagulation (EC) process

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### ABSTRACT

An experimental study was conducted under continuous flow conditions to evaluate some of the factors influencing contaminant removal by electrocoagulation (EC). A bench-scale simulation of drinking water treatment was done by adding a filtration column after a rectangular EC reactor. Contaminant removal efficiency was determined for voltages ranging from 10 to 25 V and a comparative study was done with distilled water and tap water for two contaminants: nitrate and arsenic(V). Maximum removal efficiency was 84% for nitrate at 25 V and 75% for arsenic(V) at 20 V. No significant difference in contaminant removal was observed in tap water versus distilled water. Increase in initial As(V) concentration from 1 ppm to 2 ppm resulted in a 10% increase in removal efficiency. Turbidity in the EC reactor effluent was 52 NTU and had to be filtered to achieve acceptable levels of final turbidity (5 NTU) at steady-state. The flow regime in the continuous flow reactor was also evaluated in a tracer study to determine whether it is a plug flow reactor (PFR) or constantly stirred tank reactor (CSTR) and the results show that this reactor was close to an ideal CSTR, i.e., it was fairly well-mixed.

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### 1. Introduction

Electrocoagulation is the process of destabilizing suspended, emulsified, or dissolved contaminants in an aqueous medium by introducing an electric current into the medium. In its simplest form, an electrocoagulation reactor is made up of an electrolytic cell with one anode and one cathode. The conductive metal plates are commonly known as 'sacrificial electrodes' and may be of the same or different materials (anode and cathode). When an electric current is passed, the anode dissolves, resulting in the production of metal cations. These cations then spontaneously form polymeric metal hydroxide species in solution, similar to coagulant salts like alum and ferric chloride in conventional chemical coagulation (CC). These cations and other charged polymeric metal hydroxide species can bring about neutralization of negatively charged particles. Once they are neutralized, the particles can bind together to form aggregates or flocs, resulting in pollutant removal. An additional process that takes place in electrocoagulation, besides coagulation, flocculation and settling is electroflotation. Due to the formation of gas bubbles at both electrodes, electroflotation occurs which results in additional removal of contaminants that are not removed in CC.

The EC process has proven very effective in removing contaminants from water and wastewater and is characterized by reduced sludge production, no requirement for additional chemicals and ease of operation [1,2].

In recent years, electrocoagulation (EC) has attracted great attention as an eco-friendly, efficient and cost-effective process. Removal efficiencies as high as 99% through EC have been reported for the removal of contaminants. Lack of a systematic approach to electrocoagulation reactor design and operation and the issue of electrode reliability (particularly passivation of electrodes over time) have limited its implementation. However, recent technical improvements combined with a growing need for small-scale decentralized water and wastewater treatment facilities have led to a re-evaluation of electrocoagulation.

EC requires simple equipment, and forms less sludge which is readily settleable and easy to dewater as it is primarily composed of metallic oxides and hydroxides. Flocs formed by EC are similar to chemical flocs except that EC flocs are larger, contain less bound water and are more stable. Hence, they can be separated faster by settling and filtration. As the process avoids use of chemicals therefore, no excess chemicals are used. The EC process is controlled electrically with no moving parts, thus requiring less maintenance. It can also be used in rural areas along with solar energy where electricity is not available. EC has some demerits like replacement of electrodes at regular interval due to their dissolution, high cost of electricity, anode passivation, and gelatinous hydroxides which may tend to solubilize in some cases. These problems

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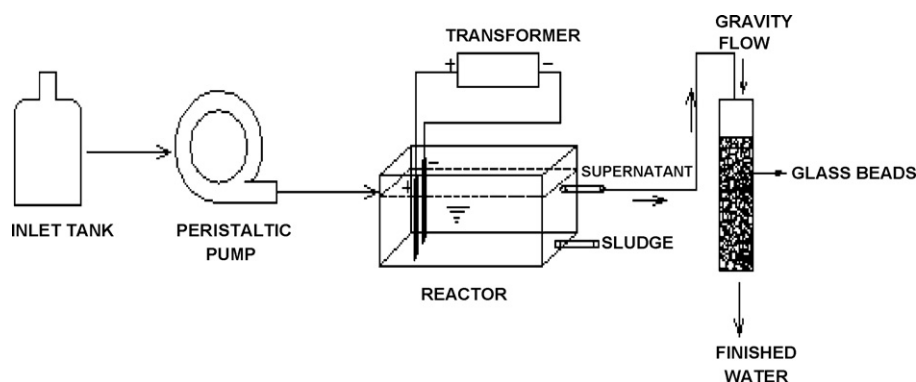


Fig. 1. Schematic diagram of experimental set-up.

can be overcome by frequent current reversal, and use of solar power [2].

### 1.1. Nitrate

Pollution of ground and surface waters by nitrates is a widespread and serious global problem. Nitrates are generally considered non-toxic, and are toxic only to infants, where ingestion of nitrates can result in the blue-baby syndrome—'methemoglobinemia'. Nitrates are essential nutrients and contribute to the eutrophication of standing surface water bodies. Heavy utilization of artificial fertilizers has resulted in very high nitrate concentrations in ground waters. Concentrations as high as 469 mg/L were reported in the High Plains aquifer in the Mid-West region of the USA [3]. In India, concentrations as high as 273 mg/L have been reported in the Najafgarh block of the Delhi region [4], while 166 mg/L were reported in Bithore in the Kanpur region [5]. The maximum allowable concentration of nitrate as  $\text{NO}_3^-$  in drinking water according to IS 10500 is 45 mg/L (15 mg/L for infants) [6].

Current treatment methods for the removal of contaminants like nitrates are ion exchange, precipitation, biological treatment, reverse osmosis and electrochemical processes. Nitrate ions can be removed from water using anion exchange resins. A high excess of regenerant, most notably NaCl, is required to regenerate the resin periodically. The collected nitrate ends up in the waste solution resulting from the brine regeneration process. Disposal of the waste solution may create significant costs and regulatory problems. In biological treatment, microbiological contamination of water is possible and the control and the effectiveness of the method has its limitations [7].

### 1.2. Arsenic

Arsenic poisoning in humans is characterized by adverse effects on the central nervous system leading initially to coma and eventually to death. It is also believed to cause lung and skin cancer, gastrointestinal disorders, muscular weakness, loss of appetite, etc. Arsenite [As(III)] and arsenate [As(V)] are highly toxic inorganic arsenic species of which arsenite is 25–60 times more toxic. The maximum allowable concentration of arsenic in drinking water is 50 ppb according to the IS 10500 [6].

Arsenic is conventionally removed by chemical techniques such as treatment with lime, alum, ferric hydroxide, ferric sulphate, sodium sulphate. Other processes used with varying degrees of success include adsorption on activated carbon and ion exchange [8]. Conventional methods of arsenic removal generate considerable quantities of secondary pollutants such as sludge, which pose serious environmental problems. While a few research groups have

reported on As removal by electrocoagulation [8–13], 5 out of the 6 studies found in the literature were based on batch processes. Only one study was found that reported electrocoagulation as a remediation tool for wastewater containing arsenic in a continuous process [11].

The objective of the present study was to evaluate the extent of removal of contaminants like nitrate, and arsenic(V) from drinking water using electrocoagulation in a continuous flow reactor. Various factors that can influence removal efficiencies were evaluated and included a range of voltage settings (10–25 V), distilled water versus tap water spiked with the contaminants, need for filtration after EC, and reactor flow regime, i.e., whether it is a constantly stirred tank reactor (CSTR) or plug flow reactor (PFR). Various parameters were monitored during each experimental run. The parameters monitored were pH, conductivity, turbidity and effluent concentrations of As(V) or nitrate. Sludge production and electrode consumption were also determined for each run.

## 2. Experimental procedure

### 2.1. EC reactor and experimental setup

A continuous flow EC reactor was made in the lab from Perspex sheet with dimensions of 36 cm × 12 cm × 11.5 cm and a length to width ratio of 3. It was run at a flow rate of 2 L/h, and a design hydraulic retention time of 2 h. The electrodes used in the EC process were mild steel of size 14.5 cm × 2.5 cm × 0.1 cm with immersion depth of 9 cm and distance of 7 cm between them. A schematic of the experimental setup for the EC reactor is shown in Fig. 1. Applied potential was varied using a transformer which converts AC to DC and the current was measured with multimeters during the experimental run. Effluent from the EC reactor was passed through a filter column of diameter 20 mm filled with 6 mm glass beads to remove suspended solids/turbidity. The finished water was analyzed for pH, turbidity and total dissolved solids (TDS) after filtration.

Coagulation, flocculation, settling and flotation were taking place within the single reactor unit. The supernatant (or clarified water or reactor effluent) above the settled floc was sampled at the reactor end opposite the electrode end of the reactor and filtered using cellulose nitrate paper of diameter 47 mm with a nominal pore size of 0.45  $\mu\text{m}$  (GFF grade paper, Whatman India). pH and turbidity were monitored during the course of each experiment in the influent and effluent. Effluent samples were taken at different times during the experiment and the experiment was continued until steady-state concentrations were achieved. In general, each experimental run lasted 8 h. All effluent samples were analyzed for pH, turbidity and As(V) or nitrate.

### 2.1.1. Nitrate

A total of seven continuous flow experiments were conducted with nitrate spiked solutions of distilled water and one experiment with tap water spiked with nitrate. Nitrate solutions were prepared using potassium nitrate ( $\text{KNO}_3$ ) and the initial nitrate concentrations in all experiments were 300 ppm. Four experimental runs were conducted at different voltages: 10 V, 15 V, 20 V and 25 V. In one experimental run, effluent from the EC reactor was passed through a filter column of diameter 20 mm filled with 6 mm glass beads to remove suspended solids. The filtered water was analyzed for turbidity after filtration. Additionally, a tracer study was done with nitrate (at an initial concentration of 100 ppm) to analyze the hydraulics of the EC reactor, i.e., the degree of mixing or whether it is closer to an ideal PFR or a CSTR, and to determine the mean hydraulic residence time in the reactor.

### 2.1.2. Arsenic

Four continuous flow experiments were conducted with As(V) spiked solutions of distilled water and one experiment with tap water spiked with As(V). As(V) solutions were prepared using sodium arsenate ( $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ ). Three experimental runs were conducted at different voltages: 10 V, 15 V, and 20 V, and the initial As(V) concentrations in all 3 experiments were 1 ppm. The effect of varying the initial As(V) concentration was studied by changing the initial concentration to 2 ppm and maintaining the voltage at 20 V. Another experiment was done at 20 V by spiking a tap water solution with 1 ppm of As(V) to compare removal efficiency in tap water versus distilled water.

## 2.2. Analytical methods

### 2.2.1. Nitrate

Concentrations of nitrate were measured at an absorbance of 229 nm as per Method 4500 [14]. A UV Spectrophotometer (Model UV2601, Beijing Rayleigh, China) was used to determine the concentrations of nitrate.

### 2.2.2. Arsenic

Arsenic concentrations in the EC supernatant were measured by the SDDC (silver diethyldithiocarbonate) Method 3500C [14]. Absorbance of the colored solution was measured at 535 nm wavelength in a Visible Spectrophotometer (GENESYS 20, USA).

### 2.2.3. pH and conductivity

A pH–conductivity meter (Cyberscan 510, Oakton Instruments, USA) was used to monitor the pH and conductivity in the EC cell.

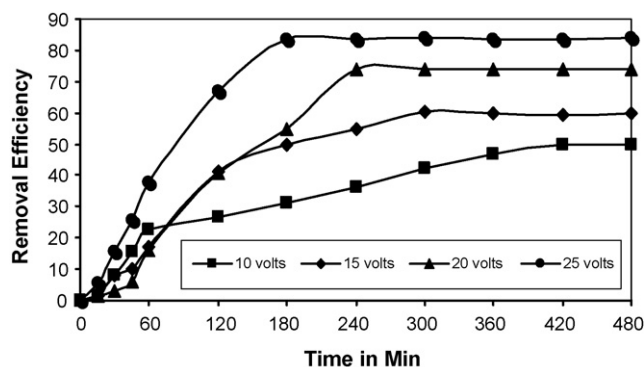
### 2.2.4. Turbidity

Turbidity meter (Orion AQUA fast II, USA) was used to measure turbidity.

**Table 1**

Summary of operating parameters for different applied potential for nitrate removal  $Q=2\text{ L/h}$ ,  $C_0=300\text{ mg/L}$ , experimental run = 8 h (these parameters were constant for all runs).

Applied voltage (V)	$C_e$ at steady-state (mg/L)	Initial pH	Final pH	Initial turbidity (NTU)	Final turbidity (NTU)	Electrode consumption (mg/L)	Sludge produced ( $\text{kg/m}^3$ )
10	150	7.2	9.49	1.5	20	25	0.26
15	120	7.1	9.5	1.3	43	28	0.28
20	78	7.2	9.48	2	45	32	0.30
25	48	7	9.5	1.8	50	36	0.35



**Fig. 2.** Removal of nitrate (%) with time at different voltages.

## 3. Results and discussion

### 3.1. Nitrate removal

#### 3.1.1. Effect of applied potential

Higher current is expected to generate larger amounts of iron, which in turn will trap more pollutant and enhance removal efficiency. Therefore, the effect of applied potential on removal efficiency was studied using the same initial nitrate concentration of 300 mg/L. At 10 V, the maximum removal efficiency was 50.3% and it took approximately 6 h to achieve steady-state. At 25 V, the maximum removal efficiency was 84% and it took 3 h to achieve steady-state. As shown in Fig. 2, increase in applied potential resulted in decrease in time required to achieve steady-state concentration and increase in percent nitrate removal. The reason may be that with increase in applied voltage, the coagulant production rate increases and the amount of iron hydroxide available in solution to form complexes and precipitate ions also increases.

From Table 1, it is evident that all the parameters increased in proportion to the applied voltage due to increase in coagulant generation. pH increased because water is converted to hydrogen gas (from protons generated) and hydroxyl ions. Electrode consumption and sludge production also increased with increase in applied voltage.

#### 3.1.2. Comparative study with tap and distilled water

A plot of nitrate removal versus time is shown in Fig. 3 for tap water and distilled water. These results demonstrate that removal is slightly higher in distilled water compared to tap water and the difference is insignificant since it is less than the variability in nitrate measurements. A more significant difference would have been attributed to the presence of other ions in tap water.

#### 3.1.3. Simulation of drinking water treatment

Since the supernatant (or effluent) from the EC reactor had high levels of turbidity, the effluent had to be filtered. Bench-scale simulation of drinking water treatment was done by adding a filtration column after the EC reactor. The filtration column consisted of glass beads of average 6 mm diameter as filter media to remove the sus-

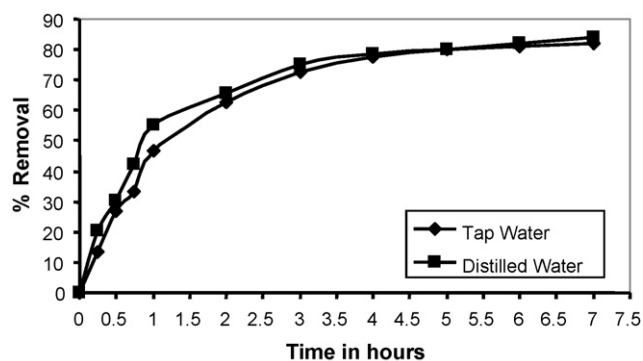


Fig. 3. Removal of nitrate with time for tap and distilled water at 25 V.

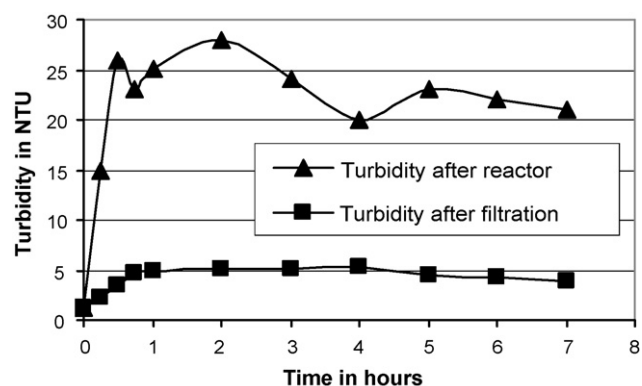


Fig. 4. Variation of turbidity with time after reactor and filtration unit for 10 V.

pendent solids coming out of the reactor. It was found that the turbidity coming out from the filtration column was less than that coming out of the EC reactor which indicates that the filter media was arresting a significant fraction of the suspended solids coming out from the reactor. A plot of turbidity versus time after EC reactor and filtration unit is shown in Fig. 4. The finished water was analyzed for water quality parameters like turbidity, pH and total dissolved solids (TDS) and it was found that turbidity and TDS met drinking water quality standards as shown in Table 2.

### 3.1.4. Tracer study

A tracer study was done with nitrate as a step input at an initial concentration of 100 ppm to study the hydraulics of a reactor, i.e. the degree of mixing in the reactor or in other words, whether it is a PFR or CSTR. It was found from the plot shown in Fig. 5 that the concentration of the exit stream is 78.5 ppm or 78.5% at a residence time of 2 h which indicates that the reactor is in between an ideal PFR and CSTR. An ideal CSTR is expected to have an exit concentration of 63 ppm while an ideal PFR is expected to have an

Table 2

Water quality parameters after EC and filtration.

Time (min)	Turbidity (NTU)	pH	TDS (mg/L)
0	1.3	6.8	350
15	2.5	7.03	375
30	3.1	8.4	400
45	4.9	9.1	387
60	5	9.33	410
120	5.1	9.42	415
180	5	9.5	420
240	4.9	9.51	405
300	4.8	9.52	389
360	4.7	9.5	400
420	4.6	9.46	418

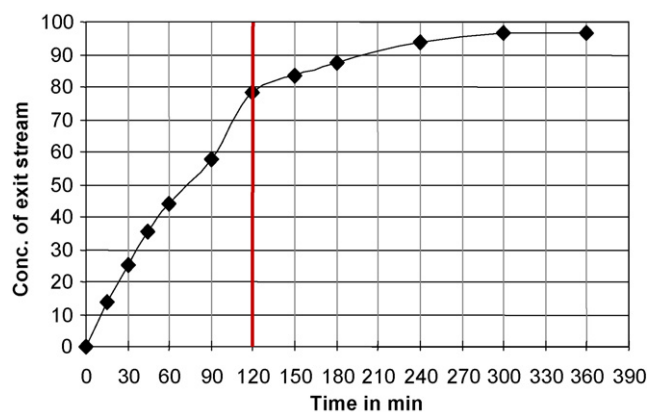


Fig. 5. Output tracer response curve from the reactor subjected to a step input of 100 mg/L of nitrate. Red line indicates design hydraulic residence time.

exit concentration of 100 ppm, respectively at the design hydraulic retention time [15]. A concentration of 78.5 ppm indicates that the reactor is close to a CSTR. The mean hydraulic residence time was calculated using the tracer data, and it was found to be 3.86 h [16].

## 3.2. As(V) removal

### 3.2.1. Effect of applied potential

The effect of applied potential on removal efficiency was studied for the same initial concentration of As(V) of 1 mg/L. It can be ascertained from Fig. 6 that as the applied potential increased, the time to achieve steady-state concentration was reduced and percent removal increased. These results are similar to that for nitrate. However in this case, it was found that the time to reach steady-state for different voltages is much less i.e., 60 min for As(V) compared to 180 min for nitrate. This may be attributed to the lower initial As(V) concentration in the reactor.

### 3.2.2. Comparative study with tap and distilled water

A plot of As(V) removal versus time is shown in Fig. 7 for tap water and distilled water. These results demonstrate that removal is slightly higher in distilled water compared to tap water and the difference is insignificant since it is less than the variability in As(V) measurements. A more significant difference would have been attributed to the presence of other ions in tap water.

### 3.2.3. Effect of varying initial concentrations

Experiments were carried out at different initial arsenic concentrations to assess the impact of initial concentration on the efficiency of electrocoagulation. It can be ascertained from Fig. 8 that the percent arsenic removal increased by 10% with an increase

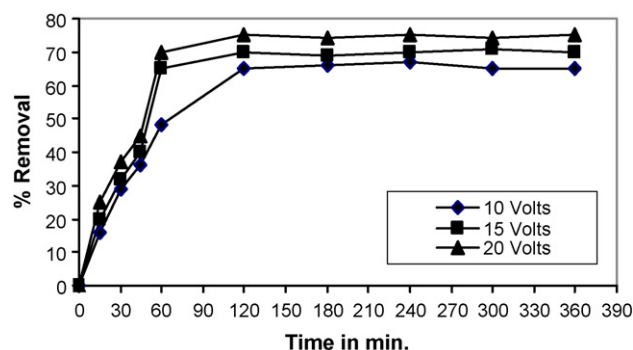


Fig. 6. Removal of As(V) with time at different volts.

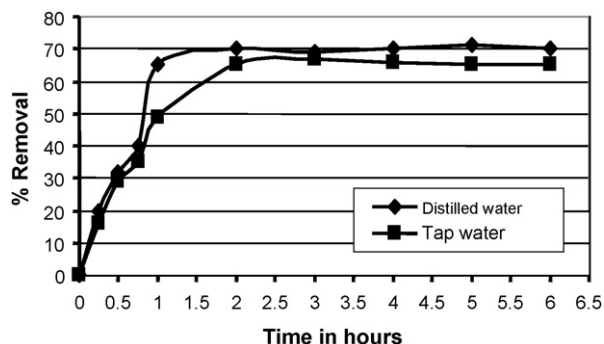


Fig. 7. Removal of As(V) from tap and distilled water at 20 V.

in the initial arsenic concentration from 1 ppm to 2 ppm. In the electrocoagulation process, the rate of arsenic removal was proportionate to the influent concentration.

### 3.3. Implications for drinking water treatment

This study has demonstrated that effective contaminant removal can be achieved in a continuous flow EC process. The EC reactor used in this study allowed coagulation, flocculation and settling to occur in a single unit. High percent contaminant removals were achieved (75% for As(V) and 84% for nitrate) in 3–6 h and there were no significant differences in removal efficiencies for tap water versus distilled water for both contaminants. The lack of difference suggests that the process is versatile in dealing with different types of source waters unlike conventional coagulation which gives very different results for different types of source waters and is very sensitive to the presence of other ions in solution [17].

While significant nitrate and As(V) removals were achieved in this study, the effluent did not meet drinking water standards under the operating conditions evaluated. Increasing applied voltage, hydraulic residence time, number and surface area of the electrodes, length:width ratio of the reactor and decreasing the distance between the electrodes are some of the changes that can be made to the process to increase its efficiency.

Other factors that were also monitored and controlled are discussed here.

#### 3.3.1. pH

The effluent pH was in the alkaline range (9.5) at steady-state for both, nitrate and As(V). This pH also corresponds to the maximum As(V) and nitrate removal efficiency. So, pH control (neutralization) after EC treatment may be required to improve removal efficiency and to bring the pH within the acceptable limits for drinking water.

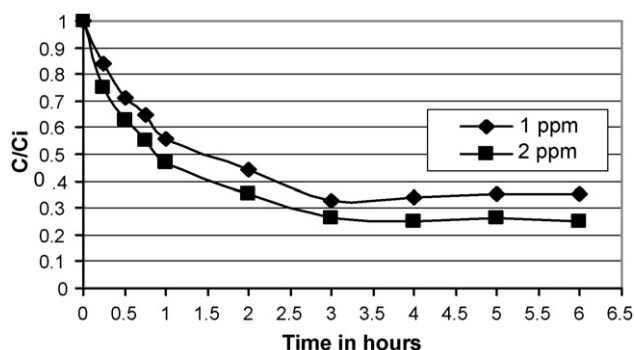


Fig. 8. Effect of initial concentration ( $C_i$ ) on As(V) removal efficiency in EC.



Fig. 9. Electrodes after the experimental run. Top portions of electrodes show initial conditions (prior to EC).

#### 3.3.2. Conductivity

During the course of each experimental run, it was observed that the conductivity of the solution increased with time as a result of which there was an increase in cell current. The increase in conductivity is attributed to the dissolution of the anode metal, i.e., elemental Fe is converted to ferrous and ferric ions and is demonstrated by the orange color (Fig. 9). The black color of the cathode is attributed to the deposition of metal oxides.

#### 3.3.3. Turbidity

During the course of the experimental run with nitrate, it was observed that turbidity increased with time reaching a maximum value of 52 NTU. Subsequently, it started decreasing and continued to do so till the end of the experimental run. The later decrease in turbidity indicates settling of floc and decrease of turbidity in the supernatant with time. If the reactor had been run for a longer time, it is likely that turbidity in the exit stream would have continued to decrease due to increase in floc formation and settling.

#### 3.3.4. Electrode consumption and cost of treatment

A major issue with the use of EC is electrode consumption and passivation. Fig. 9 shows the consumption of Fe from the anode and deposition of black Fe oxides on the cathode. Over a long period of time, this can result in a reduction in EC efficiency. One method to reduce this problem is polarity reversal. In this study, polarity of the electrodes was reversed at the beginning of each new experimental run. Operational and maintenance (O&M) costs for this study were calculated assuming that annual maintenance costs are 5% of the capital costs. O&M costs were found to be Rs.  $0.5 L^{-1}$  which is equivalent to USD  $1 m^{-3}$  at current conversion rates. It is important to note that these are actual costs for the bench-scale study conducted. A full-scale plant should be able to provide treated water at a cheaper rate given economies-of-scale. Further given the low O&M requirements, the EC process is likely to be suitable for small, decentralized systems especially in rural areas.

## 4. Conclusions

Arsenic and nitrate removal was evaluated in an EC reactor with coagulation followed by flocculation and settling; all three processes were combined in a single continuous flow reactor. In this study, some of the factors that were evaluated in a continuous flow EC reactor include: applied potential, different contaminants, different source waters and different initial concentrations of the respective contaminants. Filtration, which is required to achieve acceptable water quality in terms of turbidity, was added in one run to simulate actual conditions in drinking water treatment.

#### 4.1. Applied potential

As applied potential was increased, the time to achieve steady-state concentration decreased and percent removal of nitrate and As(V) increased. Steady-state was achieved in 60–120 min with applied voltages varying from 10 to 20 V for As(V). Maximum percent removal of As(V) at steady-state and 20 V was 75%. Steady-state was achieved for nitrate removal in 180–360 min with applied voltage varying from 10 to 25 V and the maximum percent removal was 84% at 25 V. The percent removal for As(V) and nitrate did not meet the permissible limits for drinking water in this study. However, these can be achieved by changing various operating parameters like the distance between the electrodes, applied potential, length:width ratio of the reactor, hydraulic retention time, etc.

#### 4.2. Different source waters

Two different source waters: tap and distilled waters were used for studying nitrate and As(V) removal. No significant difference in removal efficiency was noted for either contaminant. Distilled water showed slightly higher As(V) removal efficiency as compared to tap water and the small difference can be attributed to the presence of other ions in tap water. The lack of difference suggests that the process is versatile in dealing with different types of source waters unlike conventional coagulation.

#### 4.3. Different initial concentrations

Initial contaminant concentration was changed from 1 ppm to 2 ppm only for As(V) and resulted in a 10% increase in removal efficiency.

#### 4.4. Simulation of drinking water treatment

The turbidity in the effluent of the EC reactor was fairly high at about 52 NTU. Therefore, filtration was necessary to bring the effluent quality to acceptable standards. A filtration column with glass beads was added after the EC reactor to simulate actual drinking water treatment conditions. The effluent after filtration was around 5 NTU and of acceptable quality.

#### 4.5. Tracer study

The exit stream concentration of nitrate was 78.5 ppm at a design residence time of 2 h. These results show that this

reactor was close to an ideal CSTR, which is expected to have an exit stream concentration of 63.5 ppm. Based on the tracer study, the mean hydraulic residence time was found to be 3.86 h.

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