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Arsenic Removal Using Electro-coagulation: Suitable Electrode Selection

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Abstract

Arsenic contamination in groundwater and its consequences to the human health has been reported as one of the world's biggest natural groundwater calamity to the mankind. People in these affected states have chronically exposed to drinking Arsenic contaminated hand tube-wells water. One of the urgent steps towards mitigation of arsenic contamination in groundwater is supply of arsenic free or arsenic safe water. The main objective of this study was selection of suitable electrode for efficient removal of arsenic from groundwater using the method of electrocoagulation, which is a useful technique of contaminant removal. The batch mode experiments were carried out in rectangular glass cell, using different electrode combination of locally available materials such as mild steel (MS), aluminium (Al) and stainless steel (SS). The anode-cathode electrode combinations were MS-SS, MS-MS, MS-Al, Al-MS, Al-Al and Al-SS. These critical examinations had been performed under different operating condition including varying inter electrode distance, current, electrical charge loading and As (III) to As (V) ratio. The others parameters on treated water such as Fe, pH and TDS also investigated. MS anode showed 81 - 95% As removal, performed significantly well as compared to Al anode with 21 - 27% As removal for similar conditions. MS-SS was more efficient than MS-MS and MS-Al for higher arsenite to arsenate ratio. Decreasing of inter electrode distance increased arsenic removal and energy consumption. In treated water, the concentration of Fe, pH, TDS varied 0.414 - 0.057 mg/L, 7.42 - 7.60, and 391 - 413 mg/L, respectively. Fe and TDS concentration decreased whereas pH increased from initial value.

Key words: Arsenic removal; Drinking water; Electrocoagulation; Electrode; Toxicity

1. Introduction

Over the past four decades, high concentration of arsenic in drinking water has created problems in several parts of the world. Groundwater represents one of the main sources of drinking water so uncontrollable consumption has led to drilling of deeper wells (Basu *et al.,* 2014; Guzmán *et al.,* 2016). Arsenic contamination of water and wastewater has become a major social concern all over the world like India and Bangladesh which has high arsenic concentration in ground water which is otherwise safe for drinking.

With the discovery of newer sites especially in south-east Asian countries including India and Bangladesh arsenic contamination scenario has changed greatly, where people suffered from long term arsenic poisoning (Mukherjee *et al.,* 2006). From all type of source of arsenic 105 countries and territories between which 68 geogenic and 54 anthropogenic sources which covers 226.2 million are affected by arsenic (Murcott, 2012). It has also been expected that arsenic spread can be high around the world, among them part of China, Australia,

New Zealand, Afghanistan, Mali and Zambia are notable; but it has not been detected yet (Amini *et al.,* 2008).

Higher concentrations of arsenic in groundwater impacted Ganga-Brahmaputra plains in India. The aquifers in the alluvial plains are reported to be impacted, but hard rock aquifers of Karnataka and Chhattisgarh are also affected. In India permissible limit of arsenic beyond 0.05 mg/L is reported in 86 districts over 10 states and depth wise 100 m in alluvial soil. Presently, in West Bengal 11 Districts (>0.01 mg/L), 111 Blocks (>0.01mg/L) (out of which 83 blocks > 0.05 mg/L) and 16,629 habitations (>0.01mg/L) are affected by arsenic contaminated groundwater; according to census 2011, India 17.90 million of rural and 14.10 million of urban population at risk in west Bengal.

Although both inorganic and organic sources of Arsenic are present however inorganic source influences the most due to variable oxidation states and the valence depends on oxidation-reduction and pH of the water. As(V) species are found in oxidizing environment and exists as monovalent ($H_2AsO_4^-$) and divalent ($HAsO_4^{2-}$) anions in ground water and its ionic form exists at pH>3. As(III) species are found in reducing environment and exists as uncharged arsenious acid (H_3AsO_3) in ground water and it is neutral at pH < 9 and ionic at pH > 9 (Farrell *et al.,* 2001; Gomes *et al.,* 2007; Pourbaix, 1974). The valence and species of soluble Arsenic are important in assessing conventional Arsenic removal technologies. As (III) was found more than 90% in groundwater. Thus efficient conversion of Arsenic is by the chemical oxidation of As (III) to As (V) (Bora *et al.,* 2016; Kim *et al.,* 2002; Sorlini *et al.,* 2010).

Due to its high toxicity, World Health Organization (WHO) and US-EPA has lowered the maximum contaminant level (MCL) for arsenic in drinking water from 50 to 10 μg/L in 1993 and 2001, respectively. As per BIS 2012 (IS 10500:2012), the acceptable limit of Arsenic is 0.01 mg/L and the permissible limit in absence of alternate source is 0.05 mg/L (IS 10500, 2012).

Harmful health effects of arsenic depend strongly on the dose and period of exposure. Specific dermatological effects are melanosis and keratosis. Long term exposure to arsenic may also cause reproductive, neurological, cardiovascular, respiratory, hepatic, haematological, and diabetic effects in humans (National Research Council, 1999). Inorganic arsenic consumption causes bladder, lung cancer and prostate cancer (National Research Council, 2001).

There are several treatment methods deployed to remove arsenic from drinking water such as adsorption on activated alumina, activated carbon and activated bauxite, adsorption co precipitation using iron (Fe) and aluminium salts; reverse osmosis; ion exchange and oxidation followed by filtration (Viraraghavan *et al.,* 1999).

Coagulation-filtration is an effective and widespread technology useful for removal of arsenic from drinking water due to greater adsorption and discarding capabilities but they are not used for small systems due to complexities involved. Currently for small systems adsorption onto granular ferric hydroxide or granular ferric oxide or activated alumina are mainly used due to simple design and space saving purpose. The major drawback of all these techniques is that they are unable to remove As (III) effectively, however the coagulation and precipitation technique is also simple and cost effective and this makes it applicable in industries, as ferric and arsenic ions are able to reaction together (Song *et al.,* 2006). Chemical coagulant (CC) like iron salts can remove up to 90% of arsenic but it produces a large amount of sludge (Parga *et al.,* 2005).

Electro-coagulation (EC) is a prominent technology for removing arsenic but it can be used as a substitute option and resembles coagulation because the hydroxides remain unchanged, without manually adding coagulant. It has advantages of higher adsorption capacity, no manual chemical addition, media replacement, automation and easy operation and less space requirement. However EC has advantages over CC as it requires less coagulant, space, capital costs and produces less sludge (Holt *et al.,* 2002; Mills, 2000).

EC has been reported to be effective in treating arsenic from drinkable water, urban waste water (Han *et al.,* 2002), heavy-metal contaminated water, microorganisms and turbidity (Balasubramanian *et al.,* 2001; Kumar *et al.,* 2004).

In this study, an attempt has been made to remove arsenic from contaminated groundwater by electro-coagulation using different electrode combinations of locally available materials such as mild steel (MS), aluminium (Al) and Stainless Steel (SS). The anode-cathode electrode combinations are MS-SS, MS-MS, MS-Al, Al-MS, Al-Al and Al-SS. In addition an attempt has been made to identify the best suitable electrode combination. Also the process operating parameters (variables) were optimized.
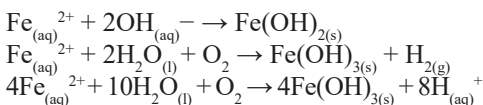
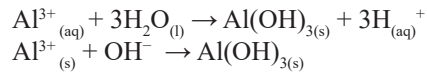## 2. Methods & Materials

2.1 Electro-coagulation mechanism

Electrocoagulation technique uses an external power source to apply potential to sacrificial electrodes (mild steel or aluminium) which undergoes oxidation to settle coagulants in their original place. The metallic cations are generated at the anode, while H_2 is produced at the cathode. In this process the gas which is generated helps the flocculated particles to float and hence this process is also called Electro-flocculation. Finally the polluting species is adsorbed onto the metal hydroxides produced (iron hydroxides or aluminium hydroxides) (Arienzo et al., 2002; Balasubramanian et al., 2001; Hansen et al., 2006; Kobya et al., 2006; Kumar et al., 2004; Parga et al., 2005) and hence removes As(III) during EC. The literature on electrochemical oxidation indicates possible generation of chlorine at the anode, which has the capacity to oxidize As(III) (Kim et al., 2002). So accordingly EC is expected to be a beneficial process in oxidizing and removing As (III) at the same time.

The electro-coagulation mechanism can be summarized as follows (Hansen et al., 2007; Kobya et al., 2011; Yousuf et al., 2001): The bulk reactions for mild steel and aluminum anodes can be written as follows;

For mild steel anode
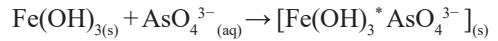$$Fe_{(aq)}^{2+} + 2OH_{(aq)}^{-} \rightarrow Fe(OH)_{2(s)}$$
$$Fe_{(aq)}^{2+} + 2H_2O_{(l)} + O_2 \rightarrow Fe(OH)_{3(s)} + H_{2(g)}$$
$$4Fe_{(aq)}^{2+} + 10H_2O_{(l)} + O_2 \rightarrow 4Fe(OH)_{3(s)} + 8H_{(aq)}^{+}$$

For aluminum anode
$$Al_{(aq)}^{3+} + 3H_2O_{(l)} \rightarrow Al(OH)_{3(s)} + 3H_{(aq)}^{+}$$
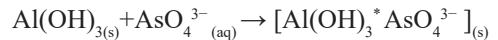$$Al_{(s)}^{3+} + OH^{-} \rightarrow Al(OH)_{3(s)}$$

The formed iron/aluminum flocs detain the arsenic present in the solution by precipitation and/or adsorption mechanism as follows;

For mild steel anode:
$$Fe(OH)_{3(s)} + AsO_4^{3-}{}_{(aq)} \rightarrow [Fe(OH)_3 {}^* AsO_4^{3-}]_{(s)}$$

For aluminum anode:
$$Al(OH)_{3(s)} + AsO_4^{3-}{}_{(aq)} \rightarrow [Al(OH)_3 {}^* AsO_4^{3-}]_{(s)}$$

The separation of suspended particles depends upon the pH of electrolyte; the separation is dominated by precipitation at low pH while adsorption dominates at high electrolyte pH (Arienzo et al., 2002).

2.2 Reagents

Stock solutions was prepared using AR grade of chemicals (Merck) and Milli-(18MUcm) for all laboratory analysis. The 100 mg As/L standard of each species were prepared from sodium m-arsenite for As (III) and sodium arsenate for As (V) from which the required amount of As (III) or As (V) was added into the water just before the experiments and made artificially contaminated.

2.3 Water sample preparation and collection

All studies were carried out with either artificially contaminated water; required amount of stock was added to arsenic free university ground water and natural contaminated water collected from source tube well. Contaminated groundwater sample was collected from a tube well (depth 23 m) located at Dhrubalok House, Purandarpur Math in Baruipur of South 24 Parganas district, latitude of N 22° 22' 22.8" and longitude of E 88° 25' 07.2" as shown in Figure 1. The water from the tube well was stored in black plastic jar for use in the laboratory. The raw water quality as per laboratory analysis is presented in Table 1.
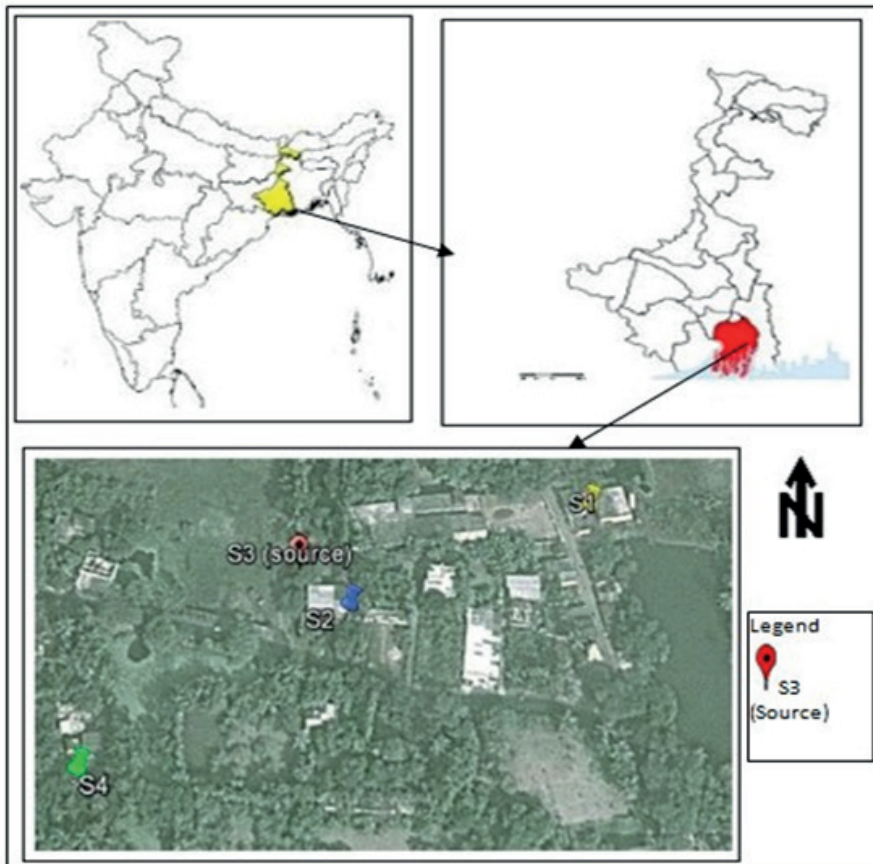
Figure 1. Tube well location of arsenic contaminated water

Table 1. Raw water quality and BIS limits

Parameter	Groundwater Baruipur	Groundwater Jadavpur University	Acceptable limit as per BIS	Maximum Permissible limit as per BIS
TDS (mg/L)	423 ± 6	1553 ± 11	500	2000
pH	7.28 ± 0.02	7.71 ± 0.07	6.5 to 8.5	6.5 to 8.5
Iron as Fe (mg/L)	2.4458 ± 0.01	1.308 ± 0.03	0.3	0.3
Arsenic as As (mg/L)	0.579 ± 0.003	BDL*	0.01	0.05

*BDL-Below Detection Limit

2.4 EC reactor

The electrical network (Figure 2) of experimental setup consisted of AC to DC converter cum voltage current regulator with AC power source, wire and tong (clip) to hold the electrodes, voltmeter (in parallel), ammeter (in series), electrode and electrolyte (contaminated water). Electrodes (submerged area 8 W × 10 H cm², thickness 1 mm) anode and cathode were connected to the positive and negative of power source respectively and hanged vertically at the middle of the cell.

2.5 Experimental procedure

All EC experiments were carried out in 7200 mL glass cell with 5000 mL spiked or natural groundwater. Then the EC was started by supplying the power and the voltage and current was noted every 5 minutes interval to calculate total energy consumption. Continuous agitation was done with magnetic stirrer at 100 rpm. Collection of sample was done using tap regulated outlet at certain minute's interval. Then the collected sample was mixed for 10 minutes at 30 rpm. Then, the whole quantity of water was allowed to stand for about 2 hour for better settling of the flocs. The total duration of EC was 30 minutes for artificially and 60 minutes for naturally contaminated water. The distance between the electrodes was varied 3cm to 5cm. The supernatants were collected in another beaker for filtration (using Whatman filter paper 42, CAT No. 1442-125, pore size 2.5 μm) and the parameters like arsenic, iron, turbidity, TDS and pH were analyzed henceforth.

2.6 Analytical procedure

An atomic absorption spectrometer (PerkinElmer AAnalyst 200), equipped with a manual hydride generator at 188.9 nm wavelength was used to determine the arsenic concentrations (detection limit of 0.1 μg/L). It is also used to determine aluminium. UV Spectrophotometer (Merck SpectroquantPharo 300) was used to analyse Iron using the 1, 10-phenanthroline method at 510 nm wavelength. These analyses were carried out based on the standard method suggested by APHA, AWWA and WEF (Rice *et al.,* 2012).
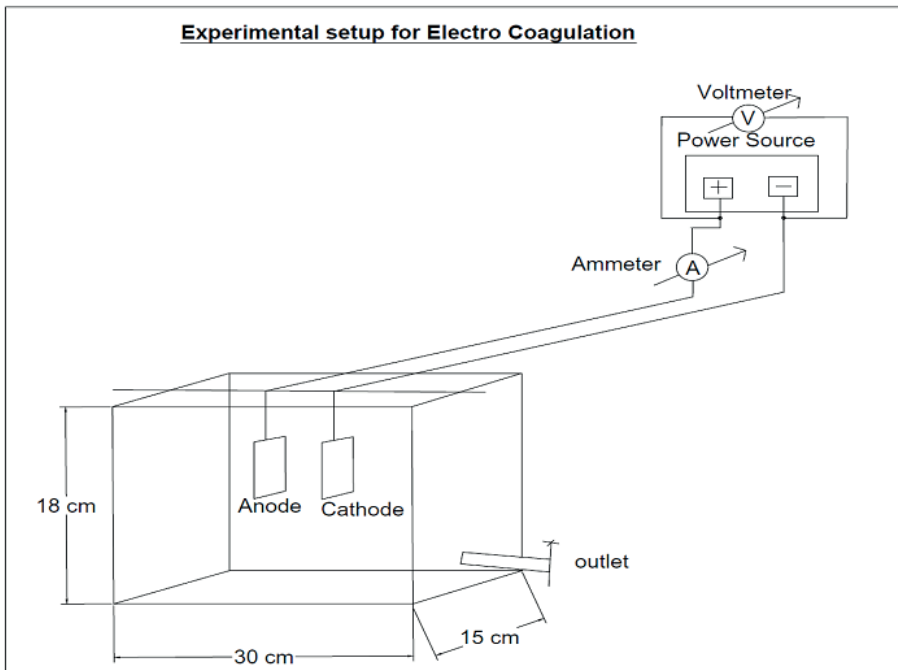

Figure 2. Network diagram of Experimental setup for electrocoagulation

3. Result and Discussion

3.1 Effects of Electrode combination

Experiments have been performed to determine the best set of electrode combinations where all other factors like initial arsenic concentration 0.598 ± 0.002 mg/L, electro-coagulation time (t_{EC}, 30 minutes) and current were maintained and hence Electrical Charge Loading (ECL) are same, only electrode combinations have been varied and response to percentage of arsenic removal are presented in Figure 3a and Figure 3b. From Figure 3a it can be observed that maximum arsenic removal after EC was 41.71% and after EC followed by filtration (i.e. overall) was 90.03% for MS-SS electrode combination. Though overall arsenic removal in case of MS-MS (88.68%) and MS-Al (84.35%) were not much less than MS-SS but removal after EC for those combinations were 27.53% and 18.92% respectively which were much less than MS-SS. In case of MS-SS maximum arsenic removal after EC indicates easy and faster agglomeration of metal hydroxide and other particle into groups, adsorption of arsenic on that, increasing the effective size and therefore settling velocities and/ or better oxidation rate. Better removal after EC decreases pollutant load on filter. MS-MS, MS-Al may be needed longer time to agglomeration-adsorption-settling for better removal of arsenic after EC. Kobya
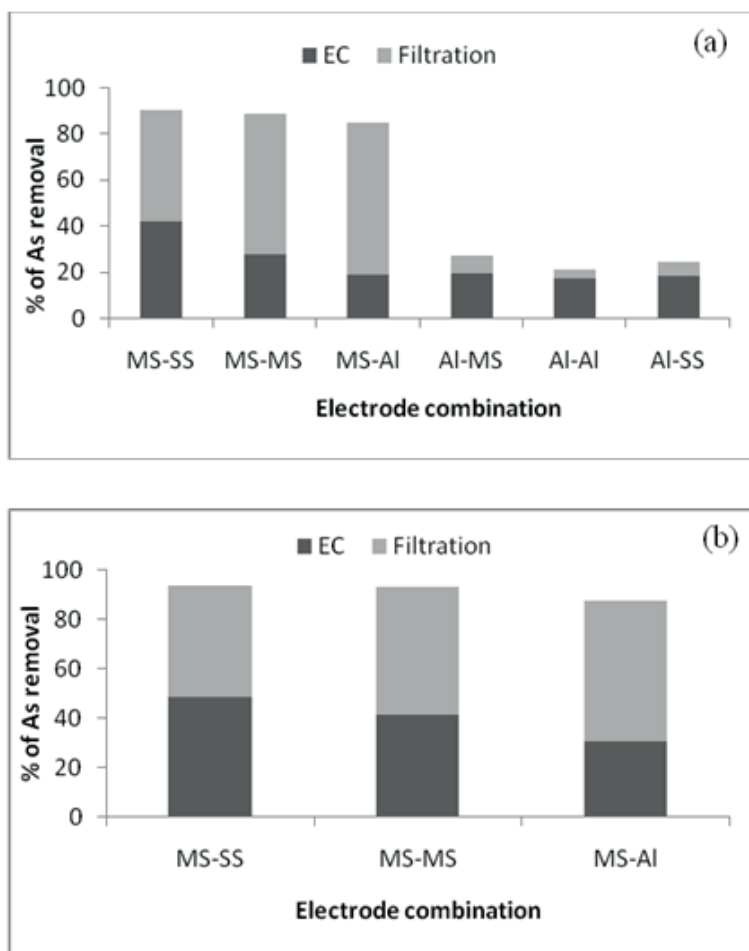

Figure 3. Influence of electrode combination on arsenic removal, (a) all MS and Al anode combination with As (III) : As (V) = 1:1 and (b) MS anode with As (III) : As (V) = 1:2

et al. (2011) observed that arsenic removal from water similar for aluminium (Al) and iron (Fe) electrode where Kumar *et al.,* (2004) was found lower arsenic removal efficiency for Al than Fe. It was observed that Al (anode) – Al (cathode) was less effective on arsenic removal than Fe or any Al and Fe mixed electrode combination (Gomes *et al.,* 2007; Kobya *et al.,* 2014). However for Al anode, it can be observed that the rate of arsenic removal is not considerable, overall removal nearly 21% to 27%. The experiments were summarized in Figure 3b carried out for further check, taking best three combination of MS anode just decreasing arsenite to arsenate ratio from 1:1 to 1:2, which are also exhibits same trends as above.

3.2 Effects of Arsenite to Arsenate ratio

Arsenite; As (III) is more toxic and difficult to remove than arsenate; As (V). Generally, for all possible techniques As (V) removed better than As (III) and oxidation is required for efficient As (III) removal (Kumar *et al.,* 2004; Nidheesh *et al.,* 2017). Since, EC itself capable of oxidation (Kumar *et al.,* 2004), without applying conventional oxidation process the effect of As (III) to As (V) ratio on total arsenic removal has been studied for best three (MS-SS, MS-MS, MS-Al) combinations already identified, initial arsenic concentration 0.598 ± 0.002 mg/L. According to the Figure 4a percentage of arsenic removal after EC increases as the ratio decreases and vice-versa. Again it
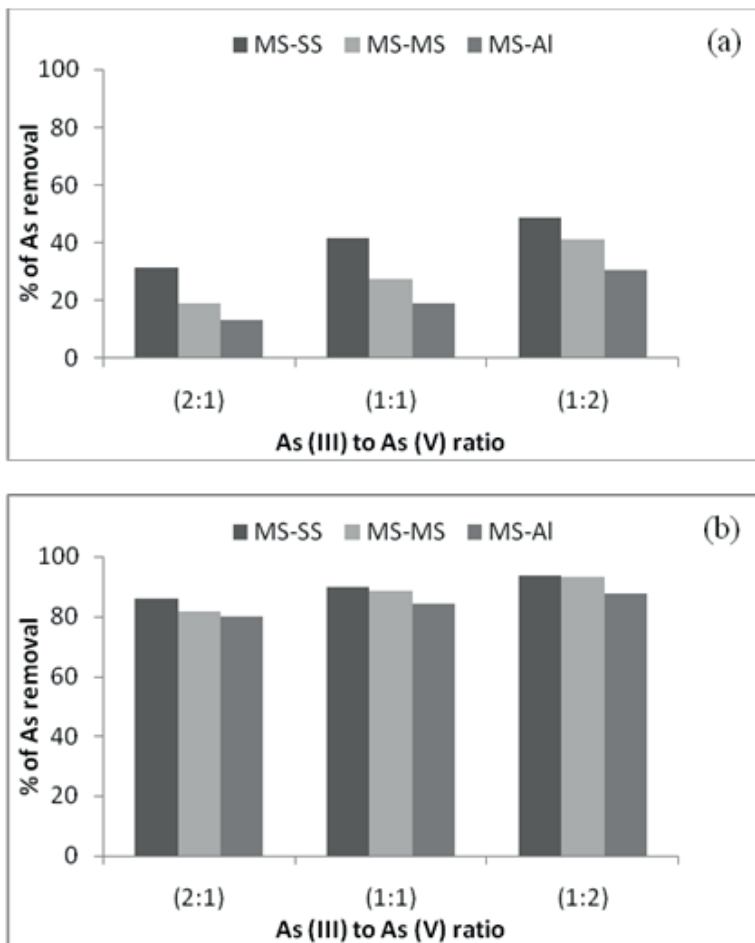
Figure 4. Influence of arsenite to arsenate ratio on arsenic removal, (a) after EC and (b) after EC followed by filtration (overall)

is maximum for MS-SS; 31.19%, 41.71% and 48.50% for ratio 2:1, 1:1 and 1:2 respectively. The difference of overall percentage of arsenic removal for MS-SS and MS-MS increases with the increase in the ratio (Figure 4b), which indicates better oxidation capacity of MS-SS during the process of EC. With respect to higher As (III) to As (V) ratio the sequence of removal efficiency is MS-SS > MS-MS > MS-Al.

3.3 Effect of inter electrode distance

Keeping the conditions like initial arsenic concentration 0.598 ± 0.002 mg/L and tEC (30 minutes) same, experiments have been performed to study the percentage of arsenic removal by varying the distance between the electrodes. From Table 2 it is observed that by decreasing the distance between the electrodes the current increases which result in the increase in ECL and anode consumption (coagulant

generation) and consequently increase the energy consumption. Hence, it can be said that the conductivity of the system increases with the decrease in inter electrode distance (IED). Thakur *et al.* (2017) had the similar observation and also noted that very small IED decreased the removal efficiency because of difficulties in circulation of contaminated water between the electrodes. As a result of the aforesaid reason the percentage of arsenic removal increases with the decrease in IED.

3.4 Effects of ECL and energy consumption

Removal of arsenic using EC mainly depends on ECL (Coulomb/L) which is directly proportional with the current and tEC and according to Faraday's law the coagulant generation increases with the increase of ECL and results in higher arsenic removal (Kumar *et al.*, 2004). From this experimental analysis Figure 6a it has been observed that as ECL
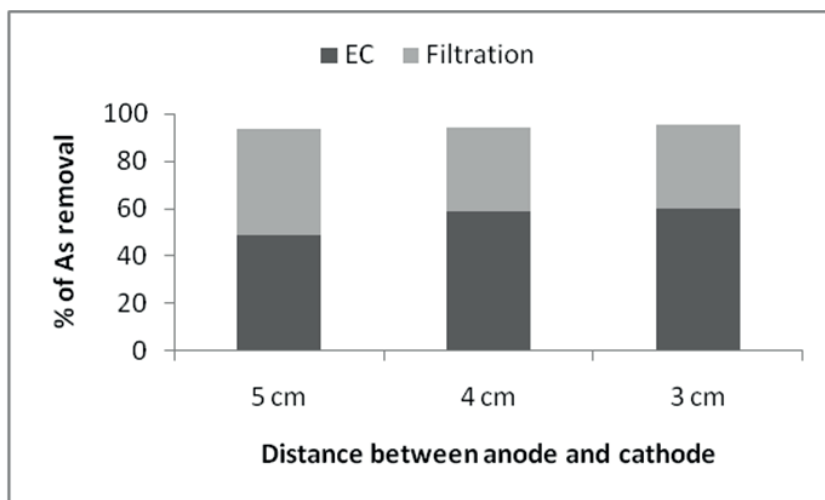

Figure 5. Influence of inter electrode distance on arsenic removal

Table 2. Variation of energy consumption and percentage of arsenic removal for the different inter electrode distance.

Inter Electrode dist. (cm)	Potential difference (Volt)	Generated Current (mA)	Electrical charge load, ECL (Coulomb)	Electrode (anode) consumption (mg)	Energy consumption (Watt-sec)	% of As Removal
5	3.04	153.66	277.22	80.23	842.73	93.65
4	2.98	161.64	292.10	84.53	870.46	94.15
3	2.87	175.26	317.15	91.78	909.14	94.98

increases the percentage of arsenic removal rate increase for any electrode combination. Similarly from Figure 6b, it has been observed that with the increase of energy consumption arsenic removal efficiency also increases. It is also noted that for a particular ECL at 150 Coulomb and energy consumption at 250 Watt-s, the removal rate is maximum for MS-SS electrode combination and the removal rate is almost same for MS-MS electrode combination. It can also be seen that for Al anode the arsenic removal rate is very slow. All the experiments have been carried out with the naturally contaminated ground water collected from tube well, in this segment
3.5 Effect on treated water; Fe, pH, TDS and Al

Influent, naturally contaminated ground water samples have been taken for this experiment which has Fe, pH and TDS value accordingly 2.446 mg/L, 7.28 and 423 mg/L (Table 1). After electro-coagulation followed by filtration effluent value of these parameters have presented graphically in Figure 7a, 7b and 7c respectively. From Figure 7a it is observed that except MS-MS all other electrodes combinations are capable to bring down Iron concentration within desirable limit (0.3 mg/L). It is found for all electrode combinations
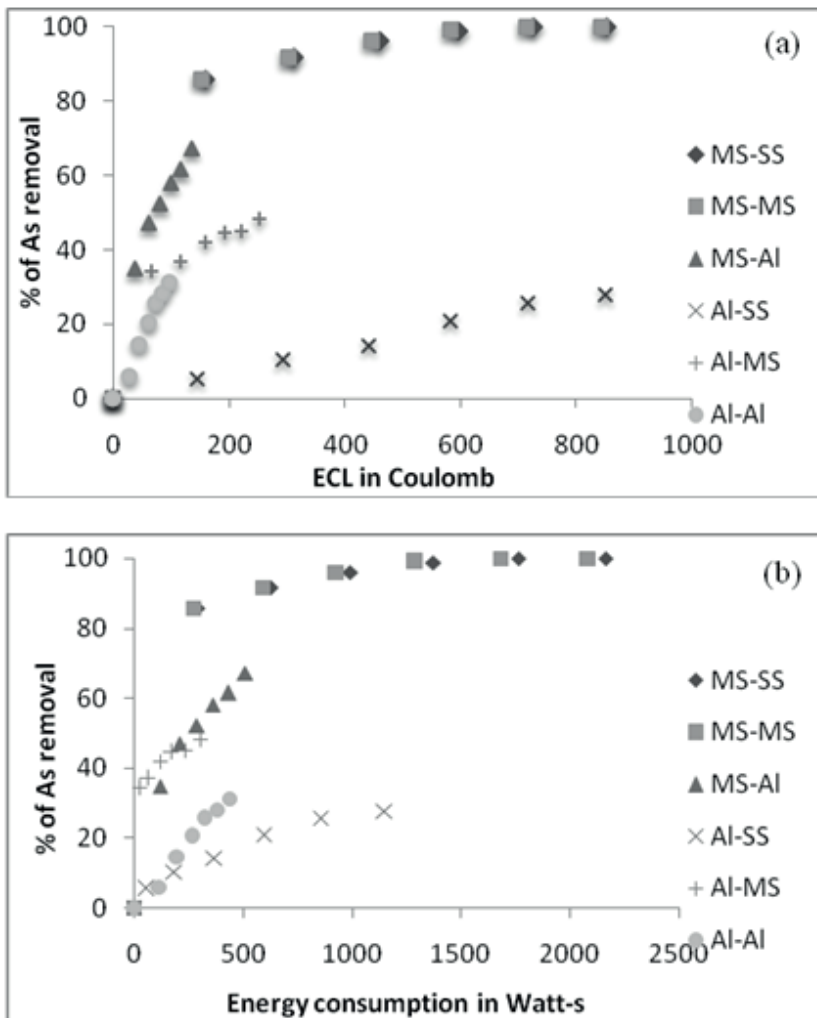

Figure 6. Influence of different electrode combinations on arsenic removal, at different (a) Electrical charge loading (ECL) and (b) Energy consumption

that pH value increases but remains within the limit (Figure 7b) and the TDS value has slightly reduced for all combination that can be observed from Figure 7c. In this context, according to Ali *et al.,* (2012) the secondary parameters like pH and TDS did not change much after treatment also iron was found below detection level. The raw water contains aluminium below detection level. However, if mild steel anode is used the aluminium content is below detection level in treated water, except aluminium cathode with mild steel anode (0.038 mg/L). Now, if aluminium anode is used, it is observed that the treated water contains 0.17 to 0.21mg/L of aluminium, which is more than acceptable limit (0.03 mg/L) (Figure 7d). Since, aluminium causes serious chronic health effects, so to remove excess aluminium from treated water further treatment is required which increases cost. So, it is recommended that aluminium electrode should not be used in this case.
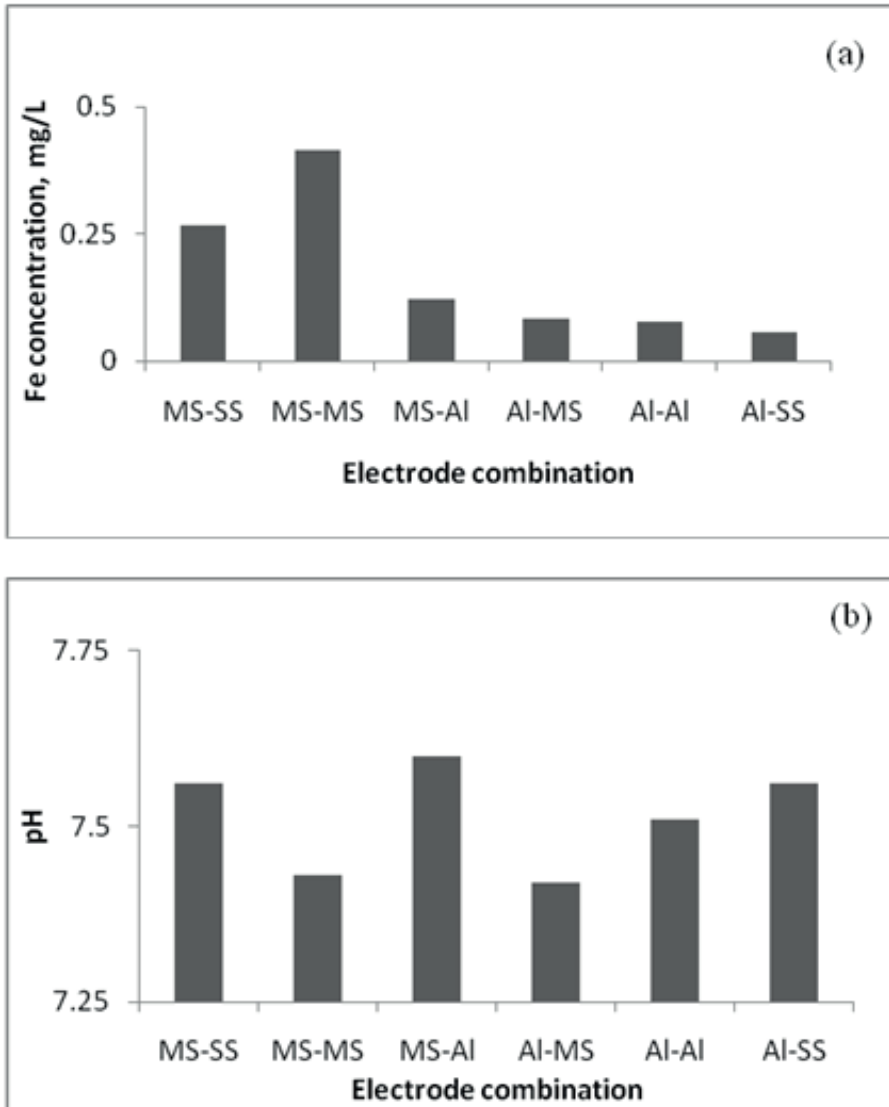
Figure 7. Influence of electrode combination ontreated water (a) iron (Fe), (b) pH, (c) TDS and (d) aluminium (Al)
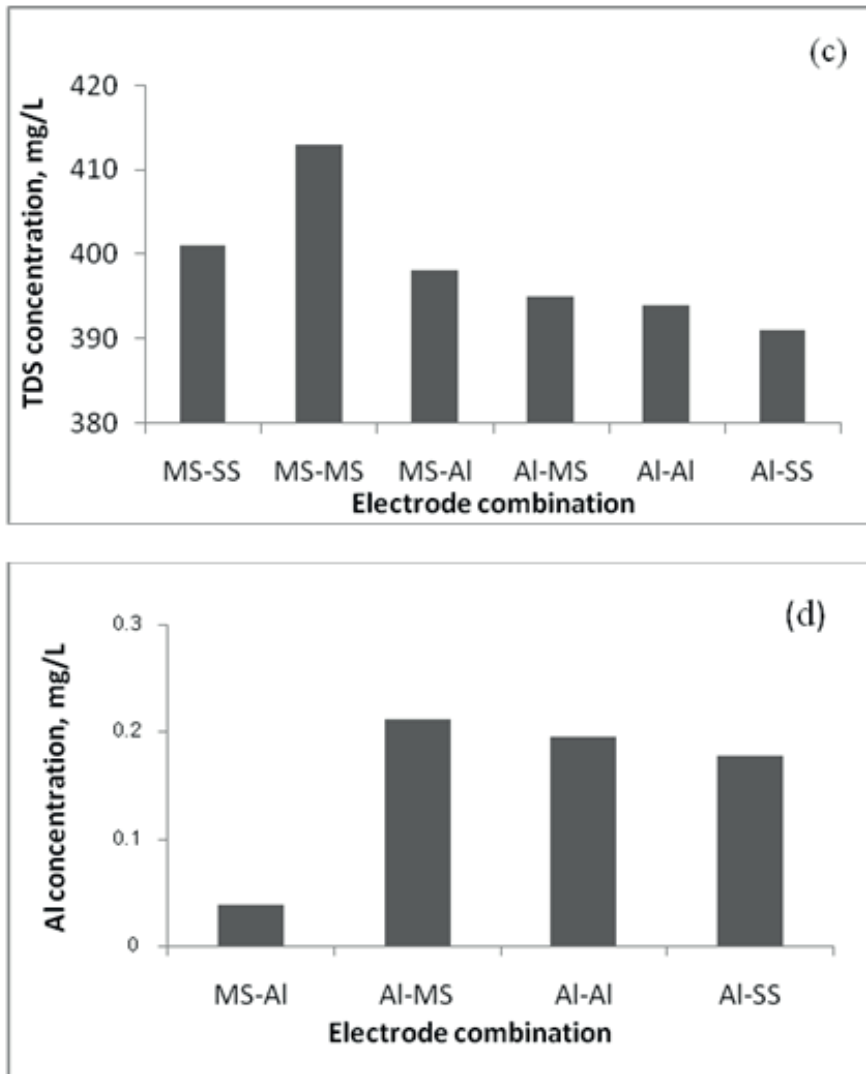
Figure 7 (Cont.) Influence of electrode combination ontreated water (a) iron (Fe), (b) pH, (c) TDS and (d) aluminium (Al)

4. Conclusion

All experiments were perpetrated to remove arsenic by electro- coagulation process under various operating conditions. The operating conditions such as electrode combination, inter electrode distance, arsenite to arsenate ratio, ECL and influent arsenic concentration have significant influence in arsenic removal. It has been observed that overall arsenic removal for MS-SS and MS-MS is almost equal and arsenic removal efficiency have been achieved from more than 81% to near about 95% depending upon the various conditions. After EC, arsenic removal is more for MS-SS which is useful to reduce contaminant loads on filter and hence filter clogging. Higher oxidation capacity is useful to remove more arsenic and MS-SS perform effectively in this aspect. It is noted that Fe, pH and TDS value for MS-SS are all within limit. So MS-SS may be selected for further development and implementation.

5. Acknowledgments

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