

# Electrocoagulation Technique For Arsenic Removal From Water - A review

<sup>1</sup>Shruthick Mamidi, <sup>2</sup>Yogesh Patil, <sup>3</sup>Ganesh kale, <sup>4</sup>Mahesh Suryawanshi, <sup>5</sup>Manisha Bagal

<sup>1</sup>Student, <sup>2</sup>Student, <sup>3</sup>Student, <sup>4</sup>Professor, <sup>5</sup>Professor,  
Department of Chemical Engineering,  
Bharati Vidyapeeth College Of Engineering, Navi Mumbai, India.

**Abstract :** Arsenic is deemed highly toxic to human beings. Over 296 million people residing in more than 100 countries have already been reported to be affected by arsenic rich groundwater. Upon long term exposure it can cause severe health issues, even short term chronic intake can cause health problems at higher concentrations. In terms of oxidation states, it exists in two common forms viz., Arsenite As (III) and arsenate As (V) in water. Not all treatment methods can remove both forms of arsenic from water effectively. Electrocoagulation is an effective technique used for arsenic removal. This review focuses on important aspects of EC reactor such as effect of operating parameters and conditions to achieve optimal performance. It also discusses factors influencing energy consumption and efficiency of the process based on recent developments.

**Keywords :** Arsenic pollution, current density, Coagulant, Electrode, operating parameters, floatation.

## 1. Introduction

Arsenic is a metalloid found on earth's crust. having atomic number 33. Naturally, it occurs in many minerals, usually in combination with sulfur and metals, but also as a pure elemental crystal. It can contaminate groundwater through rock drainage during mining or other such activities (Ramos et al., 2011).

Arsenic is a highly toxic when in water in its inorganic form. World Health Organization and and Regulatory Water Indian Standards have specified the guideline a maximum concentration limit of 10 ppb ( $\mu\text{g/L}$ ). International Agency for Research on Cancer (IARC) has also classified Arsenic in drinking water as carcinogenic stating its long term exposure can cause lung, bladder and skin cancers. Other adverse health effects which will be related to long-term ingestion of inorganic arsenic include developmental effects, diabetes, pulmonary disease, and disorder Arsenic-induced myocardial infarction, in particular, can be a significant cause of excess mortality (Rahman et al.,2011).

Considering the WHO provisional guideline value of 10  $\mu\text{g/L}$  at least 140 million people in 50 countries have been drinking water containing arsenic at levels above the safe limit. Some countries where the problem of groundwater arsenic contamination exists are : Argentina, Bangladesh, Chile, Hungary, Canada, Pakistan, China, Mexico, Taiwan, South Africa, USA, Vietnam and West Bengal (India). The South and Southeast Asian As Belt are considered the most arsenic polluted areas; these include India, Bangladesh, Nepal, Vietnam and China (Ravenscroft et al.,2009).

Until now several treatment techniques have been proposed for removing arsenic from water. Among these more frequently employed technologies are chemical coagulation, adsorption, membrane filtration, reverse osmosis, oxidation and ion exchange. However these methods have their own drawbacks such as higher operating costs, time consuming, generation of large amount of secondary pollutants (sludge), poor removal of As (III) species, higher amount of chemical reagents requirement, lower efficiency (Sudipta G et al.,2018, Sadiya A et al.,2020).Electrocoagulation is an electrolysis process in which oxidation of anodic material leads to the formation of coagulant. Instead of adding chemical coagulant externally it is generated when current is applied to the electrodes. Recently Electrocoagulation has gained a lot of attention due to its effectiveness in rapid removal of arsenic at relatively lower costs. In this paper we will discuss the technical aspects, operational features as well as difficulties encountered during arsenic removal by electrocoagulation.

## 2. Electrocoagulation reactor and its operation

### 2.1 Factors related to Electrodes

Electrocoagulation reactors needs electrodes as one of its main components. Electrodes can affect the rate of coagulant generation, flow pattern of electrolyte in case of continuous mode and thus plays an important role in reactor operation. Some of the design aspects relating to the electrodes are:

#### 2.1.1 Material of Electrodes

Electrode material oxidizes and results in the formation of coagulant. The material for electrodes should be such that upon oxidation it forms the species which has coagulative properties. Iron and aluminium best fit this criteria and are therefore commonly used. There are two sorts of electrodes : Anode and cathode. Either same material can be used for both in which case polarity of electrodes can be switched periodically or different materials can be used for both anode and cathode. For example cheaper graphite cathodes can be used which are speculated to have  $\text{O}_2$  reduction (Y.Qin et al.,2020).

Kumar et al. (2004) studied arsenic removal with a Iron, aluminium and titanium electrodes and found iron to have highest removal. Similarly Bisara et al. (2019) used air cathodes and Iron, aluminium and magnesium as anodes and found arsenic removal efficiency to be highest for Iron among Iron magnesium and aluminium anodes. However Kobya et al. (2011) found that aluminium electrodes provided slightly better performance with higher removal efficiency 95.7 % for Al and 93.5 % for Fe electrodes.

### 2.1.2 Electrode Geometry/shape

Generally plate-type, ball and rod shaped electrodes are used for electrocoagulation processes. Possibly this choice can be made based on the surface area it provides. If more surface area is provided by a certain geometry within a specific reactor volume then more metal surface will be available for dissolution. Consequently more coagulant will be generated within given time.

Koby et al. (2015) compared performance of plate and ball type electrodes where they found plate electrodes to be better.

Plate and rod type electrodes have some disadvantages namely, being time consuming in changing & maintenance and accommodating a limited number of electrodes with low surface area. However, they gave satisfactory performance (Demirbas et al.,2017).

Electrode consumption is another factor which must be considered. If it is higher more sludge will be generated which is unwanted. Overall Plate type electrodes seems to be a good choice.

### 2.1.3 Electrode Area to Volume ratio

The ratio of total surface area occupied by electrodes to the volume of water in the reactor ( $A/V$ ) is an important parameter. It affects the consumption of electrode and time for treatment. Number of electrodes to be used and their surface area can be determined for the given reactor volume with this parameter. On increasing the  $A/V$  ratio the removal time reduces but the electrode consumption and electricity consumption increases (M.Koby et al.,2015, Goren et al.,2020). Martinez-Villafane et al. (2009) found that  $0.466 \text{ cm}^{-1}$  (with four electrodes) was optimal  $A/V$  ratio in their studies having least energy consumption and for  $A/V$  ratio less than  $0.35 \text{ cm}^{-1}$  treatment time required was much higher. If the distance between the electrodes is reduced it would allow us to incorporate more electrodes and increase  $A/V$  ratio. Reduced distance between electrodes also results in increased mass transfer as a result of because of the turbulence generated. Also, it has been found that energy consumption, treatment time and electrode consumption are lower when distance between electrodes is shorter (Martinez et al., 2009, Molgora et al., 2013).

### 2.2 Mode of Operation

Similar to other any process, electrocoagulation reactors can be operated in either batch or continuous mode. Electrocoagulation is a combination of three phenomena viz, electrochemistry flotation and coagulation, which makes it difficult in regards to mathematical modelling of reactors. It is more convenient to design reactor based on experimental observations.

Batch reactors are simply cuboidal or cylindrical tanks equipped with electrodes and a power source. They are easier to operate and does not require any pumps. Therefore they are economically affordable. The sludge generated at the end of operation can be removed by passing the water through a sand filter.

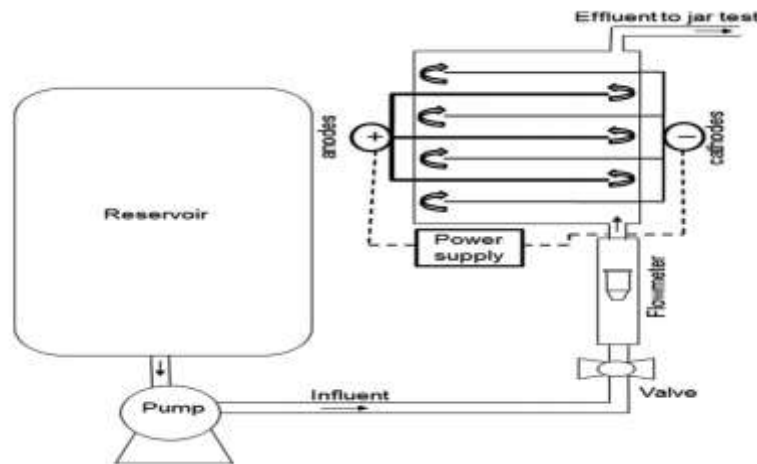


Fig. 1. Filter press type EC reactor (taken from Flores et al. (2013))

Continuous mode factors can be designed in a variety of forms. Most common being continuous tank reactors, fixed bed reactors and filter press type (plug flow) reactors. Among these, plug-flow reactors have poor performance compared to others. Flow path is created by arranging electrodes in alternated manner (Molgora et al., 2013, Flores et al., 2013). As for the packed bed reactors, anode material is packed in the form of balls or scraps at the center and water is passed through it while the cathode surrounds the packing (Demirbas et al., 2017).

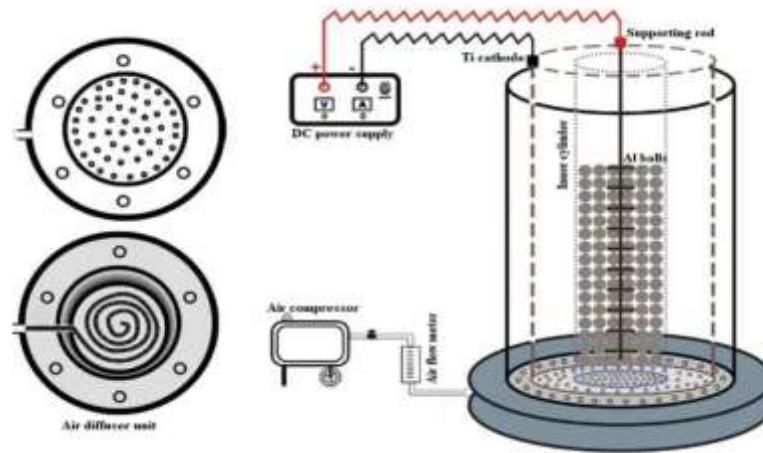


Fig. 2. Aerated packed bed reactor (taken from Goren et al. (2020))

In continuous mode flow rate becomes another significant operating parameter. Electrolyte flow allows the coagulant to be dispersed throughout the reactor utilizing it effectively. A slower inlet flow rate increases formation of stable coagulant molecules giving higher removal efficiency (Maitlo et al., 2019).

## 2.3 Modifications to the reactor

### 2.3.1 Air bubbling

Air bubbling can be provided by using diffusers or any other external means. It has two important roles in the removal process : (i) It increases the level of dissolved oxygen in water and thus enhances the oxygen of  $Fe^{2+}$  to  $Fe^{3+}$ , (ii) It enhances the turbulence in the reactor thereby promoting the flocculation and slows down the growth of surface layers on the anode (Demirbas et al., 2017, Martinez et al., 2009).

### 2.3.2 Air Cathode

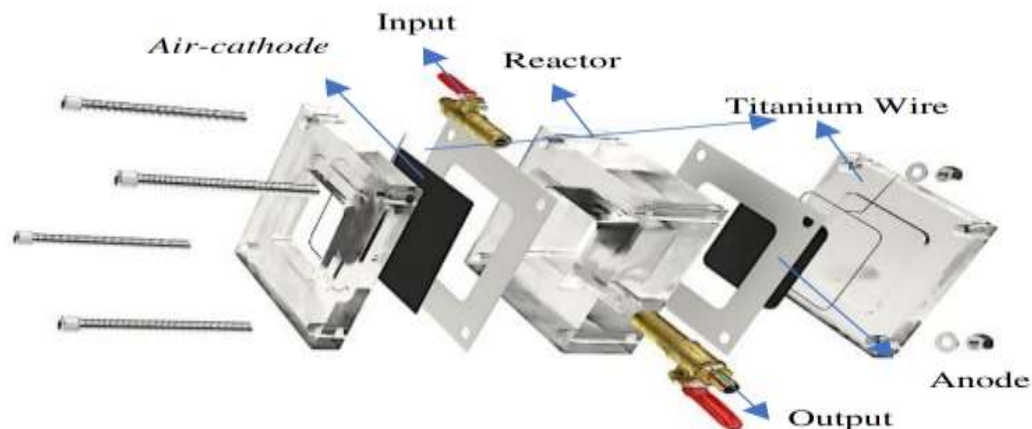


Fig 3 Air Cathode Electrocoagulation (ACEC) reactor ( taken from Bisara et al. (2020))

Electricity consumption is one of the concerning disadvantages of electrocoagulation. Air cathodes promote diffusion of atmospheric oxygen into the electrolyte. As(III) removal was found to be higher with ACEC system. Also, Air cathode electrocoagulation (ACEC) system required lower voltages for obtaining high removal efficiency. These factors make the energy consumption lower in systems with air cathodes (Bisara et al., 2020, Maitlo et al., 2019).

### 3. Operating parameters

#### 3.1 Initial pH of water

The initial pH of the water to be treated affects two processes that take place during arsenic removal. One of them is the oxidation of As(III) species to As(V) species. Other being adsorption and/or precipitation of arsenic species onto the adsorbent flocs.

For oxidation of As(III) to As(V), Fe(IV) acts as oxidizing agent and is most effectively utilized around the pH 7. Thus highest removal efficiency was obtained around this pH value (Banerji and Chaudhari, 2016).

Speciation of As(V) at various pH ranges is as follows:

$\text{AsO}_4^{3-}$  at  $\text{pH} > 12.4$

$\text{HAsO}_4^{2-}$  at  $7.2 < \text{pH} < 12.4$

$\text{H}_2\text{AsO}_4^-$  at  $3.6 < \text{pH} < 7.2$

$\text{H}_2\text{AsO}_4$  below 3.6

Beyond pH 7 adsorption of As(V) on lepidocrocite ( $\gamma\text{-FeOOH}$ ) becomes less favorable as negative charge on both As(V) and lepidocrocite increases (lepidocrocite has isoelectric pH of 7 and As(V) exists as  $\text{HAsO}_4^{2-}$  which has higher negativity) (Wan et al., 2010) and solubility of ferric hydroxide also depends on pH so it affects flocculation (Koby et al., 2011). Electrocoagulation has been found to be most effective around the pH from 5 to 8. Also, it has been observed that pH of the water increases slightly mainly because of evolution of  $\text{H}_2$  at the cathode. So, EC can act as pH neutralization step (Koby et al., 2011).

#### 3.2 Current Density

It is the most important parameter for electrocoagulation. The rate of generation of coagulant depends directly on the current density. Rate of arsenic removal increases as current density is increased (Koby et al., 2011).

According to Faraday's law current density is directly proportional to adsorbent formed as

$$C_{\text{elec, theo}} = \frac{itM}{zFV}$$

where  $C_{\text{elec, theo}}$  (kg Al or Fe electrode/m<sup>3</sup> treated potable water) is the theoretical amount of ion produced by current  $i$  (A) passed for a duration of operating time  $t$  (s),  $z$  is the number of electrons involved in the oxidation/reduction reaction;  $M$  is the atomic weight of anode material,  $F$  is the Faraday's constant (96485 C/mol) and  $V$  is the volume (m<sup>3</sup>) of the water within the vessel.

However, high energy and electrode consumption were observed at higher current densities. Consequently, operating costs increase and amount of sludge produced as well (Koby et al., 2011). Sometimes current applied is expressed in terms of current intensity (A) or charge loading as per convenience. At lower current intensity As(III) oxidation efficiency was higher (Banerji and Chaudhari, 2016). Increasing current doesn't necessarily increase the Arsenic removal accordingly. This is because at higher currents anode passivation occurs resulting in lower current efficiency.

#### 3.3 Initial Arsenic Concentration

Arsenic removal time required to reach a certain concentration (say 10 ppm) will be more for higher initial arsenic concentration for given operating conditions. It is because same amount of coagulant will be available irrespective of arsenic present in water. Arsenic removal efficiency is observed to be higher at higher arsenic concentration (Koby et al., 2011, Wan et al., 2010). Electrocoagulation has been successfully employed for concentration ranging from 36 to 100,000 ppm.

#### 3.4 Presence of other ions

This is more of a obstruction in the removal process than a parameter. When ions like silicate, phosphate, silicate, Sulphates or natural organic matter is present in water they compete with the processes involved in the arsenic removal. Some may react with Fe(II) or Fe(III) to form their respective compounds and some may compete for adsorption sites with the As(V) species. However these don't affect arsenic removal significantly if not present in large quantities (Banerji and Chaudhari, 2016, Wan et al., 2010). Table 1 shows the optimum parameters for given set of conditions.

Table 1. Summary of recent works on electrocoagulation

Reactor type	Electrode	Operating Conditions	Treatment time	Removal Efficiency	Operating Cost (\$/m <sup>3</sup> )	Reference
Continuous	Fe	C <sub>0</sub> =38±2 µg/L C.D= 48 A/m <sup>2</sup> pH=8.3 Q=12 L/h	30 min	96	0.0154	Kuan et al. (2009)
Batch	Fe-Al plate	C <sub>0</sub> = 10000 µg/L C.D=48 A/m <sup>2</sup> pH=6	10 min	99.9	-	Daniel and Prabhakara Rao (2012)
Batch	Fe	C <sub>0</sub> = 10000 µg/L C.D=52 A/m <sup>2</sup>	10 min	99.8	-	Lakshmanan et al. (2010)
Continuous	Fe	C <sub>0</sub> = 10000 pH=7.2 V=20v	60 min	75	1.0	Kumar and Goel (2010)
Continuous	Fe	C <sub>0</sub> = 10000 i = 5A pH= 7.1 Q= 30 L/min V <sub>r</sub> =100L	-	99	0.002	Parga et al. (2005)
Batch	Al Plate	C <sub>0</sub> = 150 µg/L C.D=2.5 A/m <sup>2</sup> pH=7.0	4 min	93.5	0.0073	Kobyas et al. (2011)
Batch	Fe ball	C <sub>0</sub> = 50 µg/L i = 0.05A pH=8.5	4.94 min	99	0.01	Demirbas et al. (2019)
Batch	Al ball	C <sub>0</sub> = 200 µg/L i = 0.15A pH=7.5	3 min	95	0.041	Kobyas et al. (2018)
Batch	Fe	C <sub>0</sub> = 100 µg/L pH= 7.0	16 hrs	95	0.12	Mólgora et al. (2013)
Batch	FE-Al plate	C <sub>0</sub> = 10000 µg/L C.D=47 A/m <sup>2</sup> pH=7.0	2 min	99.9	0.0782	Song et al. (2016)

## 4. Shortcomings of electrocoagulation

### 4.1 Anode Passivation

A commonly noted limitation of electrocoagulation is passivation of electrode surface via rust accumulation over continuous use for long time i.e. formation of surface layer. These macroscopic surface layers of oxidized Fe (mostly  $\text{Fe}_3\text{O}_4$ ) formed on electrodes influence long term performance of EC treatment. After several hours of continuous operation the thickness of this layer increases. It offers additional resistance to the transport of ions from the electrode and inhibits rusting of metal. Arsenic removal efficiency of field systems operated for long time has been reported to be upto 5 times less than freshly operated laboratory system [Case M Genuchten et al., 2016, S.Mullar et al., 2019, Siva R et al., 2020).

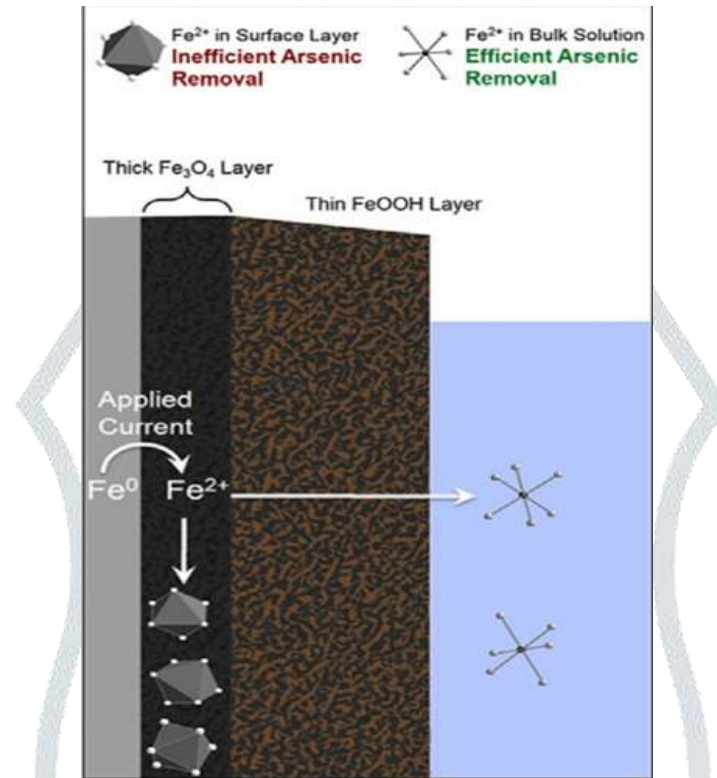


Fig. 4. Surface layer formation on electrode ( taken from Case M. van Genuchten et al. (2016))

The formation of this surface layer must be minimised to enhance performance of the EC systems. Some ways to reduce anode passivation are : (i) Reversing the polarity of electrodes frequently after set intervals of your time in order that surface layer won't form on just one electrode, (ii) Mechanical removal of surface layer at regular intervals, (iii) Modifying electrode placement in so that fluid motion aids transport of Fe(II) ions or using some mechanics that enhances turbulence, (iv) Exposing electrodes to intense ultrasonic vibrations which might limit buildup of surface layer (Case M Genuchten et al., 2016, S.Mullar et al., 2019, Siva R et al., 2020).

### 4.2 Sludge Generation

Although there's lesser generation of sludge compared to other treatment processes it still poses a drag to the environment. The sludge produced in an EC reactor passed the Toxicity Characteristic Leaching Procedure (TCLP) i.e. it's not considered hazardous waste as per U.S. standards (Amrose et al., 2013 a).

Amount and characteristics of sludge produced depends on characteristics of water and settleable solids. Higher amount of sludge is produced when current density and treatment time are higher (Kobyta et al., 2011). Analysis of X - ray diffraction showed the presence of magnetite, goethite, lepidocrocite, iron hydroxide, iron arsenate and hydrogen arsenate hydrate (Parga et al., 2013). For disposal of sludge Banerjee and Chakraborty (2005) suggested stabilization of arsenic laden sludge in briquettes, concrete and cement-sand mortars upto 40 % by volume and found it to satisfy Indian standards. This stabilized concrete can be utilised for roadway construction (Amrose et al., 2013 b).

### 4.3 Electricity Consumption

The major disadvantage of electrocoagulation is electricity consumption. It contributes the fore most to the entire energy consumption and a big part in operating costs. It is obvious that with increase in treatment time and current density energy consumption would increase. Contribution of 4 factors to the energy consumption was analysed by J. F. Martinez- Villafane and found to have their contribution in the order as follows : Distance between electrodes > Liquid motion driving mode > A/V Ratio > Current density. Contrary to expectations current density contributes least among these to the energy consumption. These factors must be considered to lower the operating costs. Limiting anode passivation and increasing current efficiency would scale back electricity consumption.

## 5. Conclusion

Electrocoagulation is a promising technology for removal of arsenic from water. It is simple, cost-effective and environment friendly. From the literature it is seen apparent that removal efficiencies as high 99.9 % can be achieved and arsenic concentration can be brought down upto 1 µg/L with electrocoagulation. In this review we have discussed several aspects which will be helpful while designing an energy efficient reactor and selecting operating parameters for treating water based on its characteristics. Effect of Electrode parameters and operating conditions on the process have also been briefly discussed. Lastly a few shortcomings of electrocoagulation and possible countermeasures on them are discussed. Electrocoagulation has high electricity consumption and which restricts its application to large scale applications. Thus further studies required are to make the process more viable. Also this techniques needs further exploration so that it can be employed for pollutants other than arsenic and various wastewaters can be treated.

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