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Removal of hardness from drinking water by electrocoagulation using iron electrodes

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ABSTRACT

In the present study laboratory studies are carried out to examine the removal of hardness by electrocoagulation process from drinking water using iron rod electrodes in batch mode. The purpose of this study was to investigate the efficiency of EC process in removal of water hardness through iron-rod electrodes in varying conditions. Experimental water sample was taken from water distribution network of local area Pune City, Maharashtra state, India. The indices for calcium and total hardness removal in pH (4.0, 7.0, and 10.0), electrical potential of 12 and 24 V and reaction times of 5 minutes to 30 minutes were measured. The maximum efficiency of hardness removal which was obtained at pH 10.0, voltage of 12 and reaction time of 60 min are equal to 98.5% and 97.5% for calcium and total hardness, respectively. Final pH of remained solution has also increased which rises with acidic pH and decreases in alcoholic pH, so the results demonstrate the direct effect of pH, potential difference and reaction time on hardness removal using EC process.

Key words: Electrocoagulation, Hardness removal, Iron electrodes, Reaction time

1. INTRODUCTION

Water is elixir of life and available for human consumption limited and likewise, available drinking water has been reduced because of the pollution created naturally and due to anthropogenic activities. Amongst water quality parameters, hardness is generated due to carbonates of calcium, magnesium and in lower traces; aluminum, iron and other bivalent and trivalent. Furthermore, water hardness is a vital parameter in industrial water consumption in manufacturing of high-quality products [1]. Hard water causes many problems in domestic and industrial consumptions like scale

formation in hot water pipes, kitchen devices, water supply facilities, boilers, cooling towers, membrane clogging, declining efficiency of heat exchangers and reaction to the soap and formation of hard foam [2]. Moreover, soft water is preferred to be hard enough to prevent Nephritis. Hence, water hardness is one of the compounds which has to be removed and its process is called water softening [2]. WHO Recommendation for drinking water's hardness is based on maximum 500 mg/l calcium carbonate [3]. Numerous technologies which need adding chemicals for water softening, are chemical precipitation and ion exchange and those which do not need to add chemicals; include reverse osmosis, electro-dialysis, nano-filtration, crystallization, distillation and evaporation [4, 5]. These techniques have some problems such as increased sludge, permanent water hardness, water salts like sodium, annual high operation costs, sediment formation on membrane, which require an effluent post treatment and disposal of residual sludge [2, 6]. Recently, growing demand for high quality water has justified the development of modern and low cost technologies for hard and very hard water softening [6]. One of these techniques is electrochemical technology such as electrocoagulation process (EC), which is being used for the removal of ions, organic matters, colloidal and suspended particles, dyes, surfactants, oil and heavy metals from aqueous environments [7–11]. This procedure has a broader potential to improve the of faults other water softening equipments. Electrocoagulation process involves three stages; coagulant formation through dissolution of metal ions of anode reactor electrode, destabilization of pollutants, suspended particles and de-emulsification, and aggregation of instable phases and floc-forming [12]. Destabilization of pollutants, suspended particles and de-emulsification mechanism can be established through dispersed double layer compression, ion neutralization species existing in water and wastewaters, and flocs and sludge forming [9]. In this study, iron electrodes have been used in electrocoagulation (EC) process. Following equations, illustrate EC process in relation to iron electrode [10]:

1) Anode Fe_(s) \rightarrow Fe²⁺_(aq) + 2e⁻ Fe²⁺_(aq) + 2OH⁻_(aq) \rightarrow Fe (OH) _{2(s)} 2) Cathode 2H₂O₍₁₎ + 2e⁻ \rightarrow H_{2 (g)} + 2OH⁻_(aq) 3) Overall Fe_(s) + 2H₂O₍₁₎ \rightarrow Fe (OH) _{2(s)} + H_{2 (g)}

From iron electrodes, Ferro ions are released into the solution through electrolytic oxidation of anode electrode and produce metal hydroxides after reacting with hydroxide monomer and polymer ions which completely depend on pH of the solution [10, 12]. Flocs generated through EC process are much larger than chemical flocs and contain more stable acid duric and less water bonds [24]. Gases produced through anode and cathode electrodes during electrolysis cause flotation and mixing gives better pollutants removal [12]. Contamination of the water caused by the application of heavy metals in the atmosphere has created alarm around the world [23]. EC process, compared with other techniques, enjoys some advantages like robust design, less resistance time, no chemicals, low sludge production, sludge stability, suitable sedimentation of sludge, dewatering and environmental compatibility [7, 9, 13]. The objective of the present study was to investigate EC process efficiency to remove hardness from water using rod-iron electrodes as a substitute for other water softening methods and determining optimal current density and pH.

2. MATERIALS AND METHODS

A laboratory experiments were performed in a batch EC reactor. Water samples were taken from local water distribution system near Pune, Maharashtra, India. Characteristics of water are shown in Table no. 1. Electrocoagulation reactor setup shown in Fig. 1 which include power supply (alternating current transformer to direct current) and iron-rod electrodes with a diameter of 2 mm connected at a distance of 2 cm into a glass beaker having volume of 1.5 L [7]. Electrodes were connected to the power supply in a monopolar and parallel arrangement which consume less energy than the series arrangement. For precise observations, each electrode was connected to positive and negative poles directly and alternately [14]; and magnetic stirrer used for mixing with needle for a speed 375 rpm. Water sample hardness rate was measured using EDTA Titrimetric method based on techniques mentioned in the standard methods of water and wastewater experiment [3]. pH of the sample was adjusted using sulphuric acid and normal sodium hydroxide; having water samples of different pHs (4, 7, and 10) under three voltages (12 and 24) and three reaction periods were tested: 5, 10, 30, and 60 min. Samples were chosen (25 mL) from the middle of the reactor using pipettes. Then, all samples were passed through a membrane filter with a size of 0.45 µm in order to remove the formed flocs. Lastly, filtered samples were analyzed concerning their calcium and total hardness.

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Sr.	Parameters	Values (mg/litre)
No.		
1	Total hardness (mg/L	315
	CaCO ₃)	
2	Calcium hardness (mg/L	140
	CaCO ₃)	
3	Phenolphthalein alkalinity	45
	(mg/L CaCO ₃)	
4	Methyl orange alkalinity	305
	(mg/L CaCO ₃)	
5	Turbidity	2 to 4 ppm
6	Temperature	28 to 30 °C
7	рН	7.13 to 7.55
8	Total Dissolved Solids	800 to 1100
	(mg/l)	
9	EC (µs/cm)	160 to 410

3. RESULTS

In this study, the efficiency of EC process using rod-iron electrodes for removing water hardness was studied as a substitute for other hardness removing techniques while using different voltages, pH and reaction times. The results of present study have been shown in Figs. 2–4. Concentration of hardness of water sample used in all experiments was constant. Fig.s 2-4 shows the efficiency of removing water hardness in electrical potentials of 12 and 24 V and pH of 4.0. As it was shown, maximum removal efficiency accomplished in voltage of 24 and reaction time of 60 min with 94.6% and 97.5% for calcium and total hardness, respectively.



Figure 1: Electrocoagulation set up: 1: DC power supply; 2 Digital Ampere meter; 3 Volt meter

Furthermore, final pH of the solution was increased from 4.0 to 10.37. Figs. 3 and 4 show the removal efficiency in the mentioned electrical potentials and in pH 7.0 and 10, respectively. In pH 7.0, the maximum removal efficiency was achieved in voltage 6 and reaction time of 60 min, which was equal to 95.5% and 95.7% for calcium and total hardness, respectively. In pH 10, the maximum removal efficiency was 97.5% and 98.5% for calcium and total hardness, respectively and reached the potential difference of 12 V and reaction time of 60 min, which considered as the maximum removal efficiency in this study. Final pH for remained solution has increased from 7 and 10 to 10.50 and 10.60 respectively, as pH does during the electrocoagulation process.

4. DISCUSSION

4.1. Effect of current density

Current density is one of the major essential factor to regulate reaction speed in EC processes as the in-situ generation of coagulants into the aqueous medium. With varying densities, the alum released into the aqueous environment decreased while the produced flocs increased, respectively; likewise, the rate of removal has also been low and high respectively [8, 9]. By increasing the density, speed and efficiency of removal process, energy and electrode consumption, the amount of produced sludge and operating costs was augmented, while, reaction time was decreased [15-17]. Thus, the effect of electrical current intensity on hardness removal from a given water was obtained in this study and observed, the removal efficiency was increased; and the current density and the time considered to be optimal for similar efficiencies were also improved, leading to decreased electrical potential differences. These results are in good agreements with the findings of arsenic removal study conducted in India, and of chromium carried out in Iran, mercury removal from water using EC with Al and Fe electrodes in France and removing humic acid from groundwater waters using EC in China [8, 18]. In this study, voltage 12 in reaction time of 60 min indicated maximum removal efficiency, i.e. 98.5% for total hardness and 97.5% for calcium hardness. Minimum removal efficiency was obtained in potential difference of 6 V; in general, voltage 12 is suggested to achieve the desired efficiency.

4.2. pH variation and its effect

Many research reported in earlier studies, that pH is a significant role in EC process and highly dependent on the pH of solution and highly impact on forming metal hydroxide and removal mechanism of ions and pollutants [8, 12, 16, 18]. Generally, pH changes during EC process depend on the type of used electrode and the primary pH [8, 12]. The value of pH increase in EC process is attributed to the formation of H₂ in cathode and aggregation of hydroxide (OH⁻) ions in the solution [16-18]. As a result, EC process could act as a pH regulator [16, 18]. In current study, three pH-ranges of 4, 7, and 10 were examined in order to explore the effect of pH on hardness removal.

Reactions for these three ranges are as follows [20]:

1) Reaction 1 (acid pH) $2Fe_{(s)} + 6H_2O_{(l)} \rightarrow O_{2(g)} + 4H_{2(g)} + 2Fe_{(OH)_{2(s)}}$ 2) Reaction 2 (neutral pH) $3Fe_{(s)} + 8H_2O_{(l)} \rightarrow Fe_{(OH)_{2(s)}} + 2Fe_{(OH)_{3(s)}} + 4H_{2(g)}$ 3) Reaction 3 (alkaline pH) $2Fe_{(s)} + 6H_2O_{(l)} \rightarrow 2Fe_{(OH)_{3(s)}} + 3H_{2(g)}$ Other reactions may be observed at high pH close 1

Other reactions may be observed at high pH close to the cathode and aggravate the precipitation of the carbonate salt on this electrode, reaction equation are as these [22]:

 $\begin{array}{l} HCO_3 + OH^- \rightarrow CO_3 \ ^{2-} + H_2O \\ CO_3 \ ^{2-} + Ca_2^+ \rightarrow CaCO_3 + CO_3 \ ^{2-} \\ CO_3 \ ^{2-} + Mg_2^+ \rightarrow MgCO_3 \end{array}$

In these reactions, produced H_2 goes upward and causes flotation and Al (OH)₃ and Al (OH)₂ precipitate. The results indicated that maximum and minimum removal efficiency was obtained in pH of 10.0 and 4.0, respectively. pHs (4.0, 7.0, and 10.0) of the remaining solution were increased to 7.37, 10.54, and 10.63 respectively, which were high in acidic pH and low in alkaline pH. Concerning iron-rod electrodes, final pH was always higher than the primary pH. The results of this study are consistent with the results of studies of potato chips wastewater [16] arsenic removal in India [18].

4.3. Effect of time

As per Faraday law, the time of electrolysis in EC process affects the rate of metal ion dissociation and released into the aqueous system [8]. Iron-rod electrodes require lesser resistance time to get a desirable removal efficiency and are economic regarding energy and electrode consumption, compared with aluminum ones [17]. The following Figures 2-4 represents, the effect of resistance time on hardness removal through EC system using rod-iron electrodes, shows good results by increasing the reaction time. So that, maximum removal efficiency was achieved in voltage of 12, pH 10 and the resistance time of 60 min which is in agreement with the results observed from arsenic removal in India [16-17], electrolytical removal of Cr⁺⁶ in UK, and chromium (VI) of synthetic solutions using EC process in Iran [17, 21, 22].



Figure 2: Removal % vs Time with 12V and 24V



Figure 3: Removal % vs Time with 12V and 24V



Figure 4: Removal % vs Time with 12V and 24V

4.4. Electrode spacing

As observed solutions lead resistance, increased distance between each couple of anode and cathode electrodes leads to enhance voltage. Due to diminished ions aggregation, hydroxide polymers, reduced rate of suspended solids and amalgamation of ions causing water hardness as well and electrostatic force, enhancing electrolyte constancy and spacing will eventually lead to decreased removal efficiency. In this study, spacing between anode and cathode electrodes was taken as 2 cm, and by reducing the distance, chemical reactions were improved due to the topical increase of concentration, and consequently, removal efficiency was increased. Present result is in line with data from other researchers like COD removal from distillery spent wash using EC with Al and Fe electrodes [17, 21] removing humic acid from groundwater waters using EC in China and also Removal of Fe (II) from tap water by electrocoagulation technique in India [18, 19].

4.5. Electrode material and effect

In electrocoagulation processes, the kind of the selected electrode material has a important effect on removal efficiency. So, it is vital important to choose a suitable type of electrode. The electrode used for drinking water treatment must be non-toxic, so, iron and aluminium electrodes were selected because of their non-toxic nature, cheap and easy accessibility [16-18, 21]. In the present study, regarding the removal of arsenic, iron-rod electrodes were 65% more efficient than its aluminum equivalents [18] and due to their high efficiencies, iron electrodes were used in this study. Initially, the color of the effluent treated by iron-rod electrodes was greenish and then changed into dark-yellow due to formation ions of ferric and ferro natures. Similar results were found treating potato-chip wastewater manufacturing factory using EC process [16, 21].

4.6. Mixing effect

Mixing is very important to generate the metal hydroxide and creates crucial role in water treatment unit which significantly affects reactions and controlling processes like sedimentation. The rod electrodes instead of flat equivalent, it was feasible to add more electrodes in the EC reactor and get a better mixing process. Furthermore, time needed to accomplish the operation problems during EC process.

5. CONCLUSION

In this study proven that EC can be used for removing hardness from water. The maximum removal has occurred at pH 10, voltage 12 and in time course of 60 min; which was 98.5% and 97.00 % for total calcium hardness and finally demonstrated the importance of direct pH, potential difference and the reaction time course on removal of hardness using EC. The overall results shows that using rod-like Fe electrodes in EC process can be effectual in removing water hardness in various situations like potential difference, acidity, reaction time and electrode type and spacing.

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