

Removal of Heavy Metals from Water and Waste Water by Electrocoagulation Process –A Review

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Abstract: Heavy metal pollution has become one of the most severe environmental problems among the different countries such as Bangladesh, Turkey, Mexico, Taiwan, Nepal, New Zealand, Vietnam, and Myanmar including India. Their non-degradable and persistence characteristic in nature make it difficult to remove. Among the various heavy metals arsenic, lead, zinc and chromium are considered to be more toxic due to their more harmful effects in living beings. Various techniques for heavy metal removal from water/wastewater are being employed such asion-exchange, adsorption, membrane filtration and coagulation/flocculation. All these techniques are not cost effective, and also produces secondary sludge, which creates environmental hazard. Electrocoagulation has gaining attention as a potential electrochemical technique for treating water/wastewater due to its versatility and environmental compatibility. The objective of the present manuscript is to review the potential of electrocoagulation process for the removal of heavy metals from water/wastewater. It is evident from the literature survey articles that electrocoagulation are the most frequently used and proficient for the treatment of heavy metal containing water/wastewater.

KEYWORDS- Heavy metals removal, Treatment technology, Electrocoagulation

1. Introduction

Heavy metals contamination in water and wastewater are severe threat to human beings in 21st century due to its toxic and non-biodegradable in nature [1]. Any metals have density greater than 5 g/cm³ is generally considered as heavy metals [2]. It has relatively high in density and poisonous in nature, even at a low concentrations, make it difficult to remove [3,4] It enters into the living beings mainly through water and food, and creates various actuate and chronic effects on human heath such as vomiting, stomach cramps, skin irritation, kidney failure as well as caner etc. Several natural and anthropogenic sources are responsible for heavy metals contamination of water and wastewater. Natural sources include weathering of rocks, volcano activity etc. whereas, anthropogenic sources include wastewater of mining, finishing, battery, paints and pigment, leather and fertilizer industries etc.[5] Many countries around the globe such as India, Bangladesh, Turkey, Mexico, Taiwan, Nepal, New Zealand, Vietnam and Myanmar affected by the presence of heavy metals in water [6]. Among the various heavy metals arsenic, lead, chromium and zinc are found normally available in wastewater, thus their removal is required.

1.1 Arsenic

Arsenic is a ubiquitous element naturally present in rocks, water and soil. It also presents in more than 250 minerals [7,8].It presents in range of 0.5 to 2.5 mg/kg in most of the rocks [8],however it varies considerably among geographic regions. Arsenic occurs in water through natural sources such as dissolution of rocks and volcano activity. The major arsenic contamination in water occurs mainly through anthropogenic sources such as burning of coal for power plants, electroplating industries, smelting of Cu, Ni, Pb, and Zn, fungicide, insecticide and organic type of pesticide that contain large amount of arsenic compounds [10].Long term consumption of arsenic contaminated water develops various skin problem such as white and dark spot, and cancer of skin, prostate,kidney, lungs and bladder [6].Therefore, the WHO sets the guidelines value of 0.01 mg/L for arsenic in drinking water.

1.2 Zinc

Zinc is essential metal for environment and human as well [11]. It naturally found in earth's crust by weathering of minerals, erosion and volcanic eruption [11, 12]. Zinc is a physically bluish- white metal which plays an important role in strengthen of

human health in lower amount, although, its high amount creates harmful effect. It enters in living beings through air, food and drinking water [13]. zinc contaminated in water and wastewater occurs mainly by the anthropogenic sources such as mining activity[14],zinc manufacturing activity [15], refinery industries, fertilizer industries, metals smelting activity[16], coal burning[17], steel production[17], agricultural activity[18], and burning of waste. Zinc deficiency causes birth defects and slow health growth. Epidermal, central nervous system, gastrointestinal, immune, skeletal and reproductive system are the organ which is mostly affected clinically by zinc scarcity [19]. Excess zinc develops vomiting [20] stomach cramps, skin irritation, kidney and liver failure [20,21].

1.3 Lead

Lead is naturally found in small amount in the earth's crust. It apparent in silver-gray [22] and bluish-gray [23] in color. It available in fossil fuel, dust and canned food [24]. Lead contamination in the environment and water increases by human activity such as mining activity, glass making, smelting of metals and formal recycling of lead, leaded petrol, paint, sand pigment and mainly by lead based batteries [25]. Lead is highly toxic and hazardous metal which affect human health in many manner such as it mainly targeted the human nerves system [24] and brain and kidney [24]. High amount of lead in human may even cause death also.

1.4 Chromium

Chromium naturally occurs in earth's crust [26]. Volcanic eruption, weathering of rocks and biological cycling are the natural sources of chromium contamination in water [26, 27]. It can exists in from of ore only and not in free elemental state [28]. Its oxidation state exists in the range of Cr^{2+} to Cr^{6+} [29]. It naturally founds in the hexavalent state in many countries such as India, USA, Africa, Finland, Philippines, New Zealand, Albania and Arizona [29]. Although, hexavalent from of chromium is much more dangerous than trivalent state [29]. Presence of high amount of chromium affects kidney, liver and nerves tissue in human body. It is a carcinogenic compound, develops stomach and respiratory cancer [28]. Heavy metals and their sources are represented in Table 1. Guideline for heavy metals concentration in drinking water are also listed in Table 2.

Metals	Sources	Reference
Arsenic	Natural sources- earth's crust, volcanic eruption, weathering of rocks	[10, 30]
	Anthropogenic sources -pesticide, insecticide, metals smelters	
	house hold product, mining activity	
Lead	Natural sources-earth's crust, volcanic eruption	[23, 25,
	Anthropogenic sources- Paint and pigment, pesticide, smoking,	40]
	automobile emission, mining, burning of coal, lead based batteries industries	
Zinc	Natural sources- earth's crust, volcanic eruption, weathering of minerals,	[11, 12, 14,
	erosion	16-18.30]
	Anthropogenic sources-refineries, brass manufacturing, metals plating, mining	
	activity, smelting activity, coal burning, steel production, agriculture activity	
Chromium	Natural sources- earth curst, volcanic eruption, weathering of rocks, biological	[26-30]
	cycling	
	Anthropogenic sources-steel industry, textile industry, electroplating, leather	
	tanning	

Table 1. Heavy metals and there sources

Name of metals	Indian standard		WHO	
	Acceptable limit	Permissible limit	Guideline value	
	(mg/L)	(mg/L)	(mg/L)	
Arsenic	0.01	0.05	0.01	
Lead	0.01	No relaxation	0.01	
Zinc	5	15	5	
Chromium	0.05	No relaxation	0.05	

Table2. Guideline for heavy metals concentration in drinking water according to Indian Standard and WHO [31, 32]

2. Removal technologies

There are various methods being employed for the removal of heavy metals from water such as ion exchange, adsorption, reverse osmosis, chemical coagulation etc. The applicability of removal methods varying according to type and amount of heavy metals present in water. The description of removal methods and their limitations are discussed below.

2.1. Reverse osmosis process

Reverse osmosis process is a physical separation method in which heavy metals are removed by passing through permeable membrane. It is a reverse process of natural osmosis. It requires high pressure for filtration with rejection of dissolved solid, metals and other pollutants. It is reported that percentage removal of lead, arsenic, zinc and chromium by reverse osmosis are 93, 95, 95 and 92 %, respectively [33,34]. Although, the removal efficiency of metals are high, but the main drawbacks of reverse osmosis process are expansive process, frequent membrane fouling, requires specific chemicals, and large amount of water is also rejected during removal. Moreover, along with the heavy metals some important minerals are also removed in reverse osmosis process.

2.2 Ion exchange method

Ion exchange process is ionic separation method in which contaminants having charge are removed by cation and anion resin from water by exchange with another non-objectionable substances. It can preciously works for industrial wastewater treatment containing low concentration of heavy metals [34,35]. Although, the removal efficiency of ion exchange process is high but the main drawbacks are: high cost and sensitivity, pH dependent and high concentration of chloride is present in treated water.

2.3 Adsorption method

Adsorption is the surface phenomenon in which active sites of adsorbent used for the removal of contaminants from water 36,37].Removal efficiency of the contaminants depends on surface area, porosity and activity of adsorbent. Adsorption process is a low cost process and easy in operation. However, the process have some drawbacks. It is a highly pH dependent process, requires pretreatment and regeneration of adsorbent, and produces several environmental issues such as disposal of exhausted adsorbent. Presence of other cations and anions also reduce the removal efficiency of contaminants.

2.4 Chemical coagulation/ flocculation

Chemical coagulation/ flocculation is a most widely method used as a pretreatment step for drinking water as well as industrial wastewater. The process involves the addition of coagulant to alter the physical state of suspended and dissolved solids and facilitate their removal by sedimentation. Coagulation is applied for removal of colloidal or suspended contaminants that do not settle out or may settle by taking a long time [34,37]. The major drawbacks of coagulation process are high requirement of chemical dose, generation of large quantity of sludge and less removal efficiency.

All the above mentioned techniques are not cost effective, and also produces secondary sludge, which creates environmental hazard. Electrocoagulation has gaining attention as a potential electrochemical technique for treating water/wastewater due to its versatility and environmental compatibility.

3. Electrocoagulation

Electrocoagulation is electro-chemical method used for the removal of contaminants and heavy metals from water and wastewater. In these method insitu coagulant is generates by applying the direct current, which helps in removal of contaminants. No addition of chemicals and high removal efficiency makes it a most promising process[34,37]. The systematic diagram of EC process is shown in Fig. 1. EC is simple and efficient method in which metal electrodes such as aluminum, iron, copper etc are generally used. The reactions occur at the anode and cathode using aluminum and iron electrode are [34,39]; At anode

$$Al(s) \rightarrow Al^{3+}(aq) + 3e^{-}$$
$$Fe(s) \rightarrow Fe^{3+}(aq) + 3e^{-}$$

At cathode

$$3H_2o + 3e^- \rightarrow \frac{3}{2}H_2 + 30H^-$$

Metal and hydroxides ions are produced at anode and cathodes, respectively get hydrolyzed in solution and form metal hydroxides [33];

$$Al^{3+} + 3OH^- \rightarrow Al(OH)_3$$

 $Fe^{3+} + 3OH^- \rightarrow Fe(OH)_2$

This in situ generate metal hydroxides act as a coagulant, have a capabilities to adsorbed contaminants, thus the high removals of these occur.

In EC process two type of electrode arrangement are normally used; monopolar and bipolar arrangement. In monopolar arrangement, two type of connection are used monopolar parallel arrangement, in this arrangement power supply is connected to each electrode in manner of anode-cathode-anode-cathode; second type is monopolar series arrangement, in this type of arrangement first and last electrode are connected with power supply and rest of others are inter connected to each other. Whereas, in bipolar parallel arrangement power supply is connected to first and last electrode rest of the electrode is sacrificial electrode and there is no inter connection between them [39,40]. The basic steps involved in EC are[41,42] (i) formation of coagulants by electrolytic corrosion of the sacrificial electrode, (ii) destabilization of the contaminants, particulate suspension and breaking of emulsions in aqueous solution, (iii) aggregation of the destabilization phase to from flocs which helps in removal of contaminants.

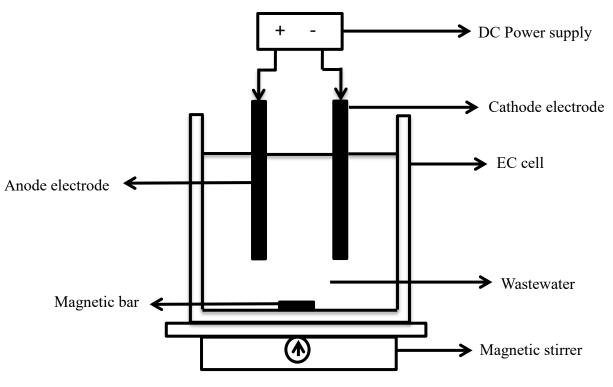


Fig. 1 Systematic diagram of electrocoagulation process [39, 40]

3.1. Factor affecting the performance of EC process

The efficiency of EC process depends on several factors such as type of electrode material, current density, inter electrode distance, solution pH, treatment time, characteristics of water etc. However, the major factors over which the removal efficiency depend are solution pH, current density, and treatment time [43,44], which are discussed below.

3.1.1. Effect of initial pH

Initial pH of the solution is an important parameter which affects the removal efficiency of contaminants. The solution pH changes during EC, thus it is usually referred to initial pH of solution. Gomes et al., 2007 observed the removal efficiency of 99.6 % of arsenic at pH value of 2.4 in case of iron electrode at 60 min treatment time. They also observed 97.5 % removal efficiency of arsenic at pH value of 6 in case of aluminum electrode at 60 min treatment time [43]. Koyba et al., 2011 found the 93.5 % removal efficiency of arsenic at pH value of 6.5 and 98.9 % at pH value of 7, in case of iron and aluminum electrode, respectively [44]. Song et al., 2016employed the response surface methodology for optimization of arsenic removal from water using hybrid Fe-Al electrode. The optimum condition for arsenic removal was found pH value of 7, current density of 0.47 A/dm², aeration of 0.32 L/min and treatment time of 20 min, at which the removal efficiency is observed 99.9 % [45]. Akbal et al., 2010 investigated the chromium removal from metal plating wastewater using iron electrocoagulation. They observed the 99.9 % removal of chromium at pH value of 3, current density of 10 mA/cm² and treatment time of 20 min [46]. Verma et al., 2013 reported the complete removal of chromium in Fe and Al EC at pH value of 4, current density of 50 mA/cm², and treatment time of 15 and 45 min for synthetic and real electroplating wastewater [47]. Dermentzis et al., 2011 studied the effect of pH on removal of chromium and zinc using aluminum EC. They observed the complete removal of chromium and zinc from synthetic and real electroplating wastewater in pH range of 4-8, current density of 40 mA/cm² and treatment time of 60 min. [48]. Hamdan et al., 2014also investigated the effect of pH on removal efficiency of chromium and zinc through continuous EC using iron electrode. The best operating condition were observed at pH 8, current density 7.61 mA/cm² and liquid flow rate of 30 mL/min at which the complete removal of chromium and 70 % of zinc was recorded [49].Koyba et

al..2015 observed the optimum conditions of pH 6.4 and treatment time of 50 min at which 99.04 % of zinc is found [50]. Mansoorian et al., 2014 also studied the effect of pH on lead and zinc removal. Low removals is observed at acidic pH (pH=3) for both the metals, which increases and found maximum at pH value of 4 and 5 for lead and zinc, respectively. It reduces further as the pH increase [51]. Kamarajet al.,2015 reported the 99.3 % of lead removal at the pH value of 7 and the removal decreased to 82.7 % when pH reduces to 3 [52]. Khosa et al., 2013compared the removal of lead, nickel and cadmium for Al and Fe electrode. They found a better lead, nickel and cadmium removal efficiency for Fe electrodes as compared to Al in case of neutral pH [53].

3.1.2. Effect of current density

Current density plays a vital role in the removal of contaminants as it decide the coagulant dose which requires for better performance of EC [54]. Gomes et al., 2007observed the > 99.6 % removal of arsenic at current density of 30 mA/m² and pH value of 4 [43]. Song et al., 2016 also found the 98.79 % removal of arsenic at current density of 0.65 A/dm². They also recorded the reduction in arsenic removal when the current density reduces [45]. Akbal et al., 2010 reported the increase in chromium removal with the increase in current density [46]. The optimum current density reported is10 mA/cm² at which complete removal of chromium observed. Dermentzis et al., 2011reported the complete removal of chromium at current density of 40 mA/cm² in case of Al electrocoagulation [48].Hamdan et al., 2014investigated the effect of current density on removal efficiency of chromium. They observed the increase in removal with increase in current density and found the maximum removal at current density of 7.61 mA/cm² [49]. Similarly, Kobya, et al., 2015 also reported the reduction in residual zinc concentration with the increase in current density [50]. They observed optimum current density of 30 A/m² for zinc removal.Bassam et al.,2012has also recorded the increase in removal of zinc with the increase in current density for Fe electrode and found the maximum removal of 96% at 25 mA/cm² of current density[55]. Mansoorian et al., 2014 studied the effect of current density on removal of lead and zinc from battery waste water [51]. Increase in lead and zinc removal is observed with the increase in current density and the optimum reported current density is 6 and 8 mA/cm² at which 96.7 and 93.8 % of lead and zinc removal occurred, respectively. Similarly, Kamaraj et al., 2015also observed the increase in removal of lead with the increase in current density [52]. When the value of current density is increased from 0.2 to 0.8 mA/cm² the lead removal efficiency also increased from 96.20 to 99.30 % [52]. The performance of EC process varies significantly with the current density. Moreover, the current density is also increases as the concentration of contaminants increase in water.

3.1.3. Effect of treatment time

Contaminants removal efficiency depend significantly on the treatment time as sufficient treatment time is requires for the contact which results further adsorption of contaminants during electrocoagulation. Gomes et al., 2007 studies showed that as the treatment time increases removal of arsenic is also increases [43]. They also reported the highest removal of arsenic in 60 min treatment time for both Al and Fe electrodes. Song et al., 2016 observed 20 min optimum time for removal of arsenic with aeration by using Fe-Al hybrid electrode [45].Verma et al.,2013 found the complete removal of chromium in 15 and 45 min of treatment time for synthetic and real electroplating wastewater, respectively[47].Akbal.et al., 2010 reported the increase in chromium removal with the increase in treatment time and found optimum treatment time of 10 min [46]. Dermentzis et al., 2011showed that the removal of zinc increase with the treatment time [48].Hamdanet al., 2014 found 99.9% removal efficiency of chromium in 10 min treatment time at air flow rate of 4 l/min [49]. Kobya et al., 2014 investigated the effect of treatment time on chromium and zinc removal [50]. Their results showed that the removals increase as the treatment time increases and found maximum at 50 min treatment time. Moreover, Khosa et al., 2013 findings suggest that removal of lead increases with the treatment time on lead and zinc removal [51]. Their findings showed that the removal increases with the passing of treatment time and achieved maximum removal at 40 min treatment time.

4. Conclusion

Heavy metal contamination of water is one of the most significant environmental issue getting attention globally. To meet the environmental regulations, an electrocoagulation process has been developed for heavy metal removal from water. It is an attractive method for the treatment of various kinds of water/wastewater, by virtue of various advantages including environmental capability, energy efficiency, versatility, selectivity, safety and cost effectiveness. In addition to that, it has a

great potential for treatment of heavy metals contaminated water. However, the removal efficiency varies significant according the contaminants type and process condition such as pH, current density and treatment time. Most of the studied reported in this review are on lab scale as well as in batch mode. Efforts should be made to perform electrocoagulation experiments at pilot plant scale to explore the possibility of using electrocoagulation in real field.

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