A Review on Removal of Heavy Metals from Water/Wastewater by Electrocoagulation Process

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Abstract: *Occurrence of toxic and recalcitrant heavy metal ions in industrial wastewater is a major environmental concern. Among the various heavy metals arsenic, chromium, lead, zinc and copper are most commonly present in wastewater. These fatal metal ions are not only hazardous in exceeding concentrations but due to the property of biomagnification, it is vital to look for the possible solutions. Electrocoagulation is emerging technology which has high heavy metals removal efficiency, easy to operate and low cost process. This review article is an effort to gather the research for finding the removal of these metals from aqueous solutions by using electrocoagulation. Studies have been complied keeping various efficiency influencing parameters such as initial pH, current density and treatment time in consideration.*

Key word: **Electrocoagulation, Heavy metal, Treatment technologies**

1. Introduction

Heavy metal pollution is one of serious environmental issue around the globe due to its toxic in nature. It is continuously increasing in environment due to industrial proliferation and urbanization **[1]**. The use of water in any activity whether it is domestic, industrial and agricultural, produces effluents which may contains these toxic pollutants. The occurrence of heavy metal in the wastewater is a major disquiet for environment. Elements which have atomic weight range from 63.5 to 200.6 and specific gravity greater than five are specified as heavy metals **[3, 4]**. Weathering and volcanic eruption are natural process responsible for heavy metal pollution. Due to the rapid development in developing countries in term of fertilizer, mining, metal plating, tanneries, pesticides, batteries and paper industries etc., a high amount of heavy metals are getting discharge in to the environment **[3, 5]**. Industrial activities mainly such as electroplating, metal surface treatment, electroless depositions, conversion coating, anodizing cleaning, etching of metals, produce large quantity of wastewater containing various heavy metal such as lead, copper, zinc, chromium, cadmium etc. Another significant source of heavy metal is the use of tin, lead and nickel plates for soldering in printed circuit board manufacturing **[5]**. Wood processing is another source where chromated copper-arsenate wood treatment produces waste containing mainly arsenic and chromium compounds **[56]**.Presence of these heavy metals in water lead to health hazards and environmental degradation. Some of the heavy metals are easily soluble in aquatic environment which can easily absorbed by fishes and vegetables and further, gets accumulate in humans if one consumes it **[60]**.Among the various heavy metals arsenic, chromium, lead, zinc and copper are the commonly present in the wastewater stream **[56]**.

2. Source of heavy metals and their consequences on human health

2.1 Arsenic

Arsenic, a ubiquitous metalloid, is associated with more than 245 mineral species. It is commonly occurs in rock formation carrying lead, copper, gold, iron hydroxides and sulfides **[8, 9]**. Volcanic eruption and other natural processes are responsible for high arsenic concentration in the environment. Excluding natural sources, arsenic contamination of soil and water also results from human exercise such as burning of fossil fuel, use of arsenic in many products, smelting of arsenic bearing minerals and disposal of waste chemicals from industries **[10]**.The concentration of arsenic in the surrounding is usually low but it is gradually increasing due to the industrialization, combustion of fossil fuel and volcanic eruption **[11]**.Another major source of arsenic is anthropogenic source, which is around 82,000 metric tons/yr worldwide **[12]**. Some of the anthropogenic

industrial sources release arsenic directly or indirectly into the environment including mining of arsenopyrite, insecticides, pesticides, weed controller, disposal of industrial and sewage materials, wood preservatives (chromated copper arsenate [CCA] chemicals), electrical waste (semiconductors) and paint products **[9]**.Arsenic is an element with a long history of toxicity **[8]**.Toxic level of this element depends on different factors such as intake rate, frequency, exposure time, oxidation state and bioavailability **[9]**.Exposure of arsenic to human beings can affects the internal organs such as kidney, lungs, liver, abnormal changes in skin and urinary bladder and produces cancer **[34]**.

2.2 Chromium

Among the heavy metal ions, chromium holds a distinct position due to its high toxic nature to biological systems. It is mainly found in chromite, which present in rock, soil, water, flora and fauna, different gases and volcanic dust. Turkey, India, South Africa and Kazakhstan are the countries where its ore chromite is mainly found. It spreads in the surrounding by several industrial actions such as electroplating, metal smelting, metallurgy, tanning, and dyestuff industries **[13, 14]**. In the aquatic environment chromium exists mainly in three states, chromium (II), chromium (III) and chromium (VI). Although, Chromium (III) is toxic in excess but an essential element for humans body because it helps in glucose metabolism. According to toxic nature chromium (VI) is more toxic than chromium (III) **[4, 15]**. Chromium (VI) disturbs or affects the human physiology, food chain and causes various health problems like skin irritation, lungs carcinoma **[16]**. Exposure of chromium (VI) to human leads to respiratory cancer **[17]**.

2.3 Lead

Lead is one of the omnipresent heavy metals which found in soil, air, sediment, surface and ground water as well as biological systems, usually in low concentration ranging from 2 to 200 ppm. Lead occurs both naturally and from human activities. Amount of lead estimated to be 3.1 x 10^{14} tones in earth crust **[18, 19]**. Natural sources of lead spread in the environment are weathering of geological materials and emissions to the atmosphere from windblown dust, volcanoes, biogenic material, sea spray, and forest fires. It is also emitted into the environment by various industrial activities such as mining and smelting, which is a primary source. According to a global inventory of trace element emissions, about 357 to 857 × 10^6kg/yr of lead is getting released in the environment by mining and smelting activities **[20]**.Concentration of lead in soil is also increasing due to land filling, incineration and composting of products made up of lead, leaded solder, glass, PVC and small lead items etc **[18]**.Lead poisoning has been accepted as one of the major public health risk mainly in developing countries **[21]**. Lead may cause pathophysiological changes in several organs including the central nervous, renal, hematopoietic and immune system **[22]**.Even low level lead exposure can strongly affect human's intelligence and also effect the function of brain **[23]**.

2.4 Zinc

Zinc is the most common metal found in the earth's crust. It is used in galvanizing process, die casting, alloys and brass in many industries. Its other compounds are also used in paints, paper and cosmetics **[26]**. Emissions of zinc to the environment occurs through mining, smelting and steel production which are the primary sources of zinc pollution **[27]**.By these activities about462 to 1380 × 10^6 kg/yr of zinc is getting released into the environment as per the global inventory of trace element emissions report **[28]**.Zinc is a toxic metal, causes many problems if intake is excessive. Zinc intake in human results vomiting, stomach cramps and nausea. Long-term exposure of zinc can leads to affect immune system, cholesterol balance and even cause infertility. On the other hand, it is an essential element which helps in growth, tissue regeneration and also for the immune system in lesser amount **[20]**.

2.5 Copper

Copper occurs naturally in rocks, soil, water, air and is an essential element in animals, plants and humans **[70]**. Sources of copper in industrial effluents are pulp, wood pulp production, paperboard mills, metal cleaning, plating baths, fertilizer industry, paints and pigments, municipal and storm water runoff etc **[71]**. Copper is essential for human life and health. But it gets toxic if continued inhalation and may leads to lung cancer **[32]**. Copper toxicity leads injury to red blood cells, lungs and can also damage liver and pancreatic functions. Long time exposure to copper may cause irritation to the mouth, nose and eyes

and may cause stomach aches, dizziness, vomiting, diarrhea and headaches **[32, 29]**. Permissible limits of heavy metal in wastewater and in drinking water are shown in Table 1.

Table1: According to Indian Standards and WHO (World health Organization) [42,43]

Heavy Metal	Permissible limits for industrial effluent discharge (in mg/l) [Indian standard,1986]			Maximum acceptable concentration in drinking water in mg/l [WHO,2017]
	Inland surface water	Public sewers	Marine coastal areas	
Arsenic	0.2	0.2	0.2	0.01
Chromium	2.0	2.0	2.0	0.05
Lead	0.1	1.0	2.0	0.01
Zinc	5.0	15	15	-
Copper	3.0	3.0	3.0	2

3. Treatment technologies for heavy metals

There are various techniques such as chemical precipitation, ion-exchange, adsorption, reverse-osmosis and electrocoagulation used for the treatment of heavy metals contaminated water **[4]**.

3.1 Chemical precipitation

In chemical precipitation process, a reagent is added which gets reacted with heavy metal ions and result information of insoluble compound or insoluble precipitates. These insoluble compounds or insoluble precipitates are further separated from water by sedimentation or filtration techniques. This treated water can be reused or discharged in the environment. Although, these method is widely used in industries but also has some limitations such as chemical used in process are corrosive; addition of treatment chemicals especially lime may increase the volume of waste sludge up to 50% and also produce H_2S fumes at low pH **[4]**.

3.2 Ion exchange

Ion exchange process is used for the separation of ions from the solution by using ion exchange resin. In this method the solution is passed through the cation and anion exchange resin, where the ions are separated based on its surface charge. These method is generally used for water and wastewater treatment in industries **[30]**.However, the method also has some drawbacks such as costly process due to resin cost; ions of the solution are replaced by sodium ions, which may cause various health effects; high operation cost as regeneration of resin is requires after regular interval etc **[25, 61]**.

3.3 Adsorption

Adsorption process is widely used in industries. Numerous natural and synthetic adsorbents are used for the water and wastewater treatment such as fly ash, neem leaf, orange peel, activated carbon, activated alumina etc **[63, 62]**. Although, the process is cost effective due to the use of natural adsorbent, but still has some disadvantages such as it is pH dependent process; adsorption capacity of the adsorbent reduces after each treatment cycle; regeneration cost of the adsorbent; requires high treatment time and low removal efficiency [**39, 64, 31**].

3.4 Reverse osmosis process

Reverse osmosis process is the opposite of natural osmosis process. It is a physical process in which membrane is used to retain the contaminants. This process requires high pressure to pass the water through the membrane **[46, 47]**. The

contaminants removal efficiency of this process is high but also suffers some limitations. It is costly process requires high initial cost as well as operation cost. It also requires specific chemical for the operation. In addition to that, large amount of water is also rejected during the process **[65, 24]**.

All the above mentioned techniques are not efficient, and produce secondary sludge which requires further treatment for disposal which makes process costly. The drawback of convention processes encourages researchers to use a new economical feasible process for treatment of heavy metals contaminated water.
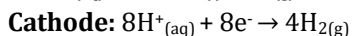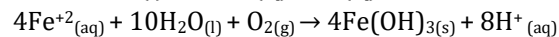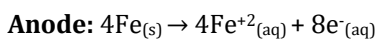
4. Electrocoagulation

Electrocoagulation is one of the most promising process gaining attention to researcher in the present era due to its high contamination removal efficiency. It is used for both the water and wastewater treatment. In electrocoagulation process, the oxidation occurs on sacrificial anode and reduction occurs at cathode in aqueous solution when current is applied. Aluminium and iron electrode material are most commonly used due to its various advantages such as availability, their low cost. The coagulant/precipitates, such as iron and aluminum hydroxides are formed insitu during the process, are non-toxic in nature, and have high contaminants removal efficiency **[33]**. Electrode material and their area, solution pH, current density and treatment time plays an important role in electrocoagulation process, whereas, presence of electrolytes and distance between electrode can also affects the process [**34**].
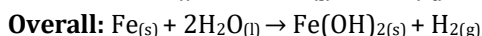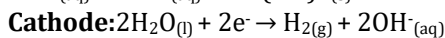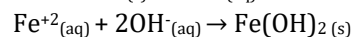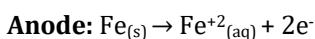
4.1 Mechanism of electrocoagulation

Mechanism of electrocoagulation is not fully known because of its complex reaction. The solution which to be treated by electrocoagulation is filled in the reactor. Electrodes of similar or dissimilar material are dipped into the solution and classified as anode and cathode. These electrodes are connected to the power source, through which the current is passed into the solution. When current is passed through aluminum and iron anodes Al^{3+} and Fe^{2+} ions, respectively, are formed. At cathode hydrogen gas and hydroxide ions are released at the same instant of time. These hydroxide ions combine with the Al^{3+} and Fe^{2+} ions in solution and formed aluminum and iron hydroxides, respectively, which act as a coagulant **[35]**. Generally, aluminum and iron are commonly used electrode in electrocoagulation. In the iron electrode, two mechanisms have been proposed.

Mechanism 1:
Anode: $4Fe_{(s)} \rightarrow 4Fe^{+2}_{(aq)} + 8e^{-}_{(aq)}$
$4Fe^{+2}_{(aq)} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 8H^{+}_{(aq)}$
Cathode: $8H^{+}_{(aq)} + 8e^{-} \rightarrow 4H_{2(g)}$
Overall: $4Fe_{(s)} + 10 H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 4H_{2(g)}$

Mechanism 2:
Anode: $Fe_{(s)} \rightarrow Fe^{+2}_{(aq)} + 2e^{-}$
$Fe^{+2}_{(aq)} + 2OH^{-}_{(aq)} \rightarrow Fe(OH)_{2\ (s)}$
Cathode: $2H_2O_{(l)} + 2e^{-} \rightarrow H_{2(g)} + 2OH^{-}_{(aq)}$
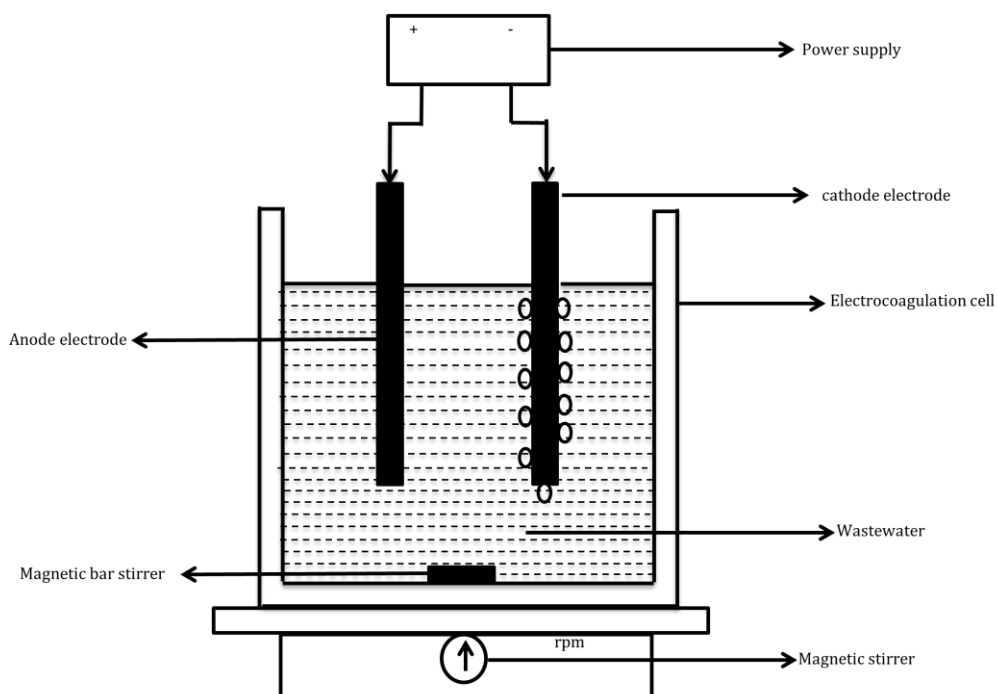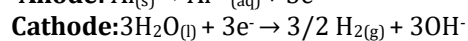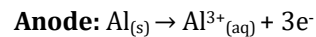**Overall:** $Fe_{(s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + H_{2(g)}$
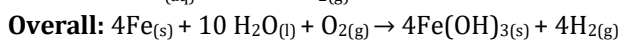
Fig .01 Systematic diagram of Electrocoagulation process
[2,35]

In case of iron electrodes various form of monomeric ions such as $Fe(OH)_3$ and polymeric hydroxyl complex such as $Fe(H_2O)_6^{3+}$,$Fe(H_2O)_5^{2+}$,$Fe(H_2O)_4(OH)_2^+$,$Fe(H_2O)_8(OH)_2^{4+}$and $Fe_2(H_2O)_6(OH)_4^{4+}$ are generate in an electrolyte system.

In the case of aluminum electrode reactions are as follows:
Anode: $Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^-$
Cathode: $3H_2O_{(l)} + 3e^- \rightarrow 3/2\ H_{2(g)} + 3OH^-$
Al^{3+}ions further react to hydroxyl ion and formed aluminum hydroxides and polyhydroxides such as $Al(H_2O)_6^{3+}$,$Al(H_2O)_5OH_2^+$, $Al(H_2O)(OH)^{2+}$etc.

4.2 Effect of various operating parameters on heavy metals removal

The heavy metals removal efficiency of electrocoagulation process depend on several operating parameters such as initial pH, current density, treatment time, conductivity of solution, electrode type and their arrangement and type of power supply in which the important parameters are initial pH, current density and treatment time.

4.2.1 Effect of Initial pH

It has been established that pH is an important parameter influencing the performance of the electrocoagulation process [7].The maximum heavy metal removal efficiency is obtained at an optimum solution pH for a specific heavy metals. It is also the well known fact that the solution pH increases during the electrocoagulation due to generation of hydroxyl ion and hydrogen gas at the cathode [66, 67].Song et al., 2016 reported the 99.9 % removal of arsenic at the optimum condition; pH value of 7, treatment time of 20 min with both Fe-Al electrodes [37].Gomes et al., 2007observed the 99.6% of arsenic removal with Fe electrode at pH value of 2.4 and 60 min treatment time. They also evaluated the arsenic removal by using the Al electrode and found 97.5 % removal at the pH value of 6 with the same treatment time of 60 min [44].Kobya et al., 2011 studied the effect of pH on arsenic removal using Fe and Al electrodes. They recorded the arsenic removal of 93.5 % at the pH value of 6.5 and 98.9 % at the pH value of 7, in case of Fe and Al electrodes, respectively [6].Hamdan et al., 2014 found the 100 % and 70 % simultaneous removal of chromium and zinc, at optimum pH of 8 [50].Akbal et al., 2010 observed 99.9 % removal of chromium at pH value of 3 in 20 min of treatment time [51].Yadav et al., 2012 investigated the effect of pH on the removal of chromium, zinc, nickel and copper. They observed the increase in the solution pH with the treatment time. They found the optimum initial pH of 2.2 at which the simultaneous removal efficiency of chromium, zinc, nickel and copper are found 100 %, 98.71 %, 69.22 % and 48.08 % respectively with Al electrode and 100 %, 75.48 %, 58.68 %, and 78.57 %, respectively with Fe electrodes in 80 min treatment time [36].Kamaraj et al., 2015 reported the optimum pH value of 7 for lead removal with the 99.3 % removal [40].Yetilmezsoy et al., 2009performed the electrocoagulation experiments for removal of lead from aqueous solution. Optimum pH was found 3.97 to achieve the complete removal of lead in 68 min treatment time [48].Dermentzis et al., 2011 reported 4-8 optimum pH range for the simultaneous complete removal of nickel, copper and zinc in treatment time of 20 min, 40 min and 50 min, respectively [49].The pH of the solution can be a key parameter for the removal of heavy metals. It varies according to type of heavy metals. Effect of pH on removal efficiency of heavy metals in electrocoagulation process is presented in Table 2.

Table 2 Effect of initial pH on removal efficiency of heavy metals

Heavy metals	Electrode material	Current density	Treatment time	pH range	Optimum pH	Removal %	References
As	Al	10 A/m2	95 min	3-11	7	98.51 %	[69]
Cr, Cu, Ni	Fe	100 A/m²	20min	5-11	3	99.9%,99.9%,98%	[51]
Pb	Al	54.9A/m²	20min	3-10	9	99%	[68]
Zn,Cu,Ni,Mn	Fe	250 A/m²	15min	3-8.95	5.68	>96% for Zn ,Cu, Ni 72.6% for Mn	[59]
Cu,Cr,Ni	Fe as anode and Al as cathode	100 A/m²	20min	3-9	3	100%	[58]

4.2.2 Effect of Current density

In electrocoagulation process current density plays a vital role as it decide the coagulant dose for the removal of heavy metals [38].Thakur et al., 2017 examined the variation of current density (10 – 60 A/m²) on the removal of arsenic by using Al electrode. The maximum removal of arsenic 98.51 % was recorded at the optimum current of 10 A/m² in 95 min treatment time [69].Kongjao et al., 2008 found the maximum 95 % removal of chromium at 22.4A/m² of current density in 20 min of treatment time [53].Adhoum et al., 2004 examined the variation of current density in the range of 0.8-4.8 A/dm² on the removal of copper, zinc and chromium. They reported the maximum simultaneous removal of copper, zinc and chromium at current density of 0.8 A/dm² in 15 min of treatment time [54].Mansoorian et al., 2014 studied the effects of current density and power supply (AC and DC) on the simultaneous removal of lead and zinc from aqueous solution by using Fe electrodes. They observed the increase in removal of both with the increase in current density. The maximum removal efficiency of lead and zinc found to 98% and 97.7%, respectively by using the direct current supply at current density of 10 mA/cm²and 40 min treatment time. In case of AC supply the maximum removal efficiency of lead and zinc found to 98.4% and 98.2%, respectively with the same operating conditions. Their results revealed that DC is more efficient than the AC.They recorded the optimum current density of 6 mA/cm²through DC supply for 97.2% and 95.5% removal of lead and zinc, respectively [39]. Kamaraj et al., 2015 evaluated the effect of current density in the range of 0.4-0.8 mA/cm²on the removal of lead in monopolar and bipolar electrode arrangement. At current density of 0.8 ma/cm², maximum 99.30 % and 99.45 % removal of lead was observed in monopolar and bipolar electrode arrangement, respectively [40]. Tezcan et al., 2015investigated the variation of current density on simultaneous removal of cadmium, copper and nickel and found maximum removal of 99.78%,99.98%,98.90%, respectively at current density of 30 mA/cm² in treatment time of 90min [52]. From the above discussion, it can be concluded that removal efficiency of heavy metals varies according to the current density. Table 3 shows the effect of current density on the removal efficiency of heavy metals.

Table 3 Effect of current density on removal efficiency of heavy metals

Heavy metals	Electrode material	pH	Treatment time	Current density range	Optimum Current density	Removal %	References
As	Al	7	95min	10-60 A/m²	10 A/m²	98.51 %	[69]
As	Fe	2.4	60 min	0.3-3 A/m²	3 A/m²	99.6%	[44]
Cr	Fe	7	20 min	15.7-24.6 A/m²	22.4 A/m²	95%	[53]
Pb	Mn– anode Fe – cathode	7	30 min	2.5-20 A/m²	15 A/m²	99%	[57]
Zn, Cu, Cr	Al	4	15 min	80-480 A/m²	80 A/m²	99.9%,99.9%,83%	[54]
Cu, Pb	Al	5	30 min	1.4-14.7 A/m²	10.2 A/m²	54.1%, 91.4%	[25]

4.2.3 Effect of treatment time

The heavy metal removal efficiency is also a function of the treatment time. The heavy metal removal efficiency increases with an increase in the treatment time, at the fixed current density, as the amount of in situ generated metal coagulant also increases with the treatment time. Thus, the increase in removal efficiency of heavy metal observed. Mansoorian et al., 2014 studied the effect of treatment time on simultaneous lead and zinc removal. They indicated that the removal increases with the increase in treatment time and reported maximum removal of lead (98 %) and zinc (97.7%) in 40 min treatment time [39].

Khosa et al., 2013 found the optimum treatment time of 10 min for the simultaneous removal of lead, nickel and cadmium using Al electrode. At these treatment time the removal efficiency of lead, nickel and cadmium were observed 90 %, 92% and 74%, respectively [41].Gomes et al., 2007also observed the increase in removal of arsenic with the treatment time.More than 98 % of arsenic found at optimum treatment time of 60min for both Al and Fe electrodes [44]. Hanay et al., 2011achieved 99.9% simultaneous removal of both copper and zinc in 5 min treatment time [55]. Kobya et al., 2010 reported 99.4 % removal of cadmium in 30 min treatment time with 30A/m^2 of current density. They also observed 99.1% of nickel removal in 80 min of treatment time with 60A/m^2 [45]. Effect of treatment time on removal efficiency of heavy metals in electrocoagulation process is presented in Table 4.

Table 4 Effect of treatment time on removal efficiency of heavy metals

Heavy metals	Electrode material	pH	Current density	Treatment time range	Optimum treatment time	Removal %	References
As	Fe	6.5	2.5A/m^2	0-15min	2.5min	94.1%	[6]
Cr	Fe	7	22.4A/m^2	0-120min	20min	95%	[53]
Pb,Zn	Fe	5	100 A/m^2	10-40min	40min	98%,97.7%	[39]
Zn, Ni,Cu,	Al	4	400 A/m^2	20-80min	20,40,50min	>97%	[49]
Cu,Zn	Al	7	150A/m^2	0-35min	5min	99.9%	[55]

5. Conclusion

The rapid urbanization and industrialization around the globe are creating high levels of heavy metal water pollution due to discharge of wastewater. The characteristic of heavy metal contaminated industrial wastewater varies according the type of industries. Similarly, the treatment process also depends on the type of heavy metals present in the wastewater. Among the various available methods electrocoagulation is one of most promising method due to its high heavy metal removal efficiency. The main factors such as pH, current density and treatment time more affect the performance of electrocoagulation process. Although, the other factors such as electrode distance, conductivity and type of power less affect the process. Moreover, studies needs to be accomplished to examine the effect of geometry and shape of the electrode to possibly improve the heavy metal removal efficiency. Presently, most of the studied reported in literature are performed at the lab scale. Efforts should be made to use electrocoagulation process in real application to explore it performance.

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