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Removal of heavy metal from electroplating wastewater using electrocoagulation: a review

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ABSTRACT

Electrocoagulation is one of the promising processes to treat a variety of wastewater including electroplating wastewater, distillery effluent, pulp, and paper mill effluent, etc. This method is well applicable to treat wastewater of chemical oxygen demand range in 1,000–20,000 mg/dm^3. In addition, the electrocoagulation process is very effective in the removal of metal or heavy metal from the wastewater depending on the nature of the metal and its concentration. However, a number of technologies such as coagulation, adsorption, precipitation, and membrane separation are also available to treat such type wastewater but in the last few decade, electrocoagulation method gains more popularity due to its versatility and environmental compatibility. The present article gives a critical and concise review of electroplating effluent towards heavy metals removal from electroplating effluent. Additionally, the role of electrocoagulation on the removal of various pollutants from different industrial wastewater including mining, textile, pulp, paper mill, distillery, chemical, paint, petroleum, and tannery are also summarized. The concept of electrocoagulation and its operating parameters are also explained in detail.

Keywords: Electrocoagulation; Heavy metal; Electroplating effluent; Organics; Operating parameter

1. Introduction

Water is mandatory for all survival including humans, animals, and plants. Nowadays due to increased industrial growth, water consumption also increases simultaneously. Industries use a large amount of fresh water for various purposes including scrubbing, coolant, carrier, raw material, etc., and discharge polluted water. This polluted water enters water sources like rivers, canals, and groundwater consequently a critical pollution problem occurs in the present scenario. Hence most of the countries have made very strict norms to discharge any industrial effluent. Additionally, currently many countries are suffering from serious water problems due to the enhancement of industrialization [1]. However, reuse and recycling of wastewater can overcome this problem up to a certain limit.

The electroplating sector is an important segment of the Indian economy that provides millions of jobs. A variety of metals like lead, chromium, zinc, and nickel are used for metal plating [2]. Hence, different types of metal used for job work are discharged from the electroplating industry. During the electroplating operation, a shrill layer of metal is coated on another metal object. The object works as cathode during the electrolytic deposition process and the anode is typically a plate of the metal to be deposited. This process is accomplished through the consumption of a large amount of fresh water. Out of these only 50% water is consumed in operation and the remaining is discharged as

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wastewater with a metal content that is commonly known as electroplating effluent (EPE). If this wastewater is directly discharged in any water receiving body without any proper treatment, then it can damage the aquatic system of the pure water stream. Furthermore, if EPE enters in groundwater source then the quality of water is fully contaminated and this polluted water can create serious health issues in term of kidney and cancer diseases [3].

There are so many established technologies available for the treatment of metal containing wastewater including coagulation [4], electrocoagulation [5], adsorption [6], precipitation [7], ion-exchange [8], membrane separation [9], etc. These methods have several limitations. For example, coagulation needs a large amount of chemicals and also generates sludge. Preparation and regeneration of adsorbent enhance the cost of the adsorption process. The membrane separation process provided poor efficiency. Precipitation and ion-exchange needs more maintenance due to large sludge production and chemical generation respectively. Apart from this, electrocoagulation process has several advantages including the least amount of chemical need, less installation, low sludge generation, and easy maintenance [10]. Therefore, the acceptability of electrocoagulation increases day by day for the treatment of different types of effluent such as textile, distillery, metal plating, etc. Table 1 presents a detailed literature review of the electrocoagulation process used for the treatment of electroplating effluent. The application of electrocoagulation on the treatment of different types of anionic impurities is given in Table 2. Furthermore, the electrocoagulation process is very effective for the removal of organics from different types of wastewaters (Table 3). However, in some cases individual electrocoagulation process does not achieve desired pollutant reduction hence the combination of electrocoagulation (EC) with other methods can be used for better pollutants reduction from wastewater (Table 4). Also, the electrocoagulation process is more effective as compared to other treatment methods as presented in Table 5.

This work serves with several novelties including a detailed literature review of electroplating effluent treatment using the electrocoagulation method, structured mechanism along with the significance of electrocoagulation. The aim of this work is to provide readers with a critical and systematic review of the treatment of electroplating effluent using electrocoagulation process. The industrial effluents such as textile, paint, distillery, and paper mill have also been incorporated towards the performance of the electrocoagulation process. The mechanism of electrocoagulation explained in detail. Additionally, the impact of different parameters such as pH, current density, electrode distance, and reaction time on the EC process also incorporated.

2. Mechanism of electrocoagulation

Electrocoagulation (EC) is a process of wastewater treatment where contaminant particles, ions such as heavy metals, and colloids are destabilizes and aggregates using an electrical charge to hold them in solution [145]. Electrocoagulation is very similar to coagulation theory except that in the coagulation process coagulant is added to water by mixing salt of iron or aluminum while in the electrocoagulation process coagulants is discharged in wastewater through the supply of electricity in the submerged sacrificial electrode (usually aluminum or iron electrode) (Fig. 1) [14]. Usually four types of theory namely (1) ionic layer compression, (2) adsorption, (3) sweep coagulation and (3) intraparticle bridging practices to understand the mechanism of electrocoagulation (Fig. 2) [146]. The detailed description is as follows:

2.1. Ionic layer compression

This theory is based on a cloud of ions present around the surface of colloids. It is a well-known fact that huge amounts of colloids are present in the water/wastewater. Colloids are always surrounded by a large number of ions that are affected by charged surface. The first layer of cations attracted by negative charged surface, is "bound" to the colloid and maybe travel with it. The remaining ions may be affected with positive charges or counter ions and come closer to the colloidal surface [94]. This type of arrangement generates a net charge that is strongest at the bound layer. When two colloids are present nearer, two types of forces act there. One is an attractive force (Vander wall force) that supports the contact and the second is a repulsive force that opposes the contact of colloids. When Vander wall force is more dominant than repulsive force, molecules cross energy barriers and particles get agglomerated [147].

2.2. Adsorption theory

According to the adsorption theory of electrocoagulation, metallic ions are released from the anode through the direct current supply to the submerged electrode. These metallic ions combined with negative ions pass by colloids consequently; the effective size of particle enhances and settles down virtue of the gravity in the form of metallic hydroxide. Following reactions occur when the iron is used as a sacrificial electrode [17];

$$\mathrm{Fe(s)} \rightarrow \mathrm{Fe^{3+}} + 3\mathrm{e^-}\,(\mathrm{anode}) \tag{1}$$

$$3\mathrm{H_2O} + 3\mathrm{e^-} \rightarrow 3/2\mathrm{H_2} + 3\mathrm{OH^-}\,(\mathrm{cathode}) \tag{2}$$

Overall

$$\mathrm{Fe^{3+}} + 3\mathrm{OH^-} \rightarrow \mathrm{Fe(OH)_3} \tag{3}$$

Hence several monomeric and polymeric species occurs during EC process via $Fe(OH)^+$, $FeOH^{2+}$, $Fe_2(OH)_2^{4+}$, $Fe(OH)_4^{5+}$, $Fe(OH)_2^0$(s) and $Fe(OH)_4^-$, etc. [136,148].

Similarly, when aluminium is used as a sacrificial electrode following anodic and cathodic reaction occurs [149]

$$\mathrm{Al} \rightarrow \mathrm{Al_3^+} + 3\mathrm{e^-}\,(\mathrm{anode}) \tag{4}$$

$$3\mathrm{H_2O} + 3\mathrm{e^-} \rightarrow 3/2\mathrm{H_2} + 3\mathrm{OH^-}\,(\mathrm{cathode}) \tag{5}$$

Table 1
Treatment of metal plating effluent using electrocoagulation process in a batch reactor

Wastewater/effluent	Electrode	EG (cm)	CD (A/m^2)	Reaction time (min)	pH^o	Metal/others contaminant	M^o (mg/dm^3)	Removal (%)	References
Electroplating effluent	Al–Al	2	200	60	6.5	Ni	Ni, 87.755–121	99.75	[11]
Copper plating effluent	Al–Al	0.6	15	15	5.0	Cu^{2+}	Cu^{2+} 200	98	[12]
Metal coating effluent	Fe–Fe	1	224	60	5.0	Pb, Cr	Pb, Cr, 3.5, 5.4	91, 91	[13]
Prepared wastewater	Al–Al	2	–	30	5.0	Pb	Pb (10–30)	99	[14]
Smelting effluent	Al–FE	1	24	60	6.0	Mn, Zn, Cd	Mn, Zn, Cd, 320, 92, 15	96, 96, 96	[15]
Metal coating industry effluent	Al–Al	0.5	–	8	3.0–9.0	Cr	Cr, 50	60	[16]
Electroplating effluent	Fe–Fe	1	80	30	8.0	Ni, Pb, Cu, Cr	Ni, Pb, Cu, Cr,13.5, 55, 4, 39	96.4, 99.5, 98, 96.2	[17]
Electroplating effluent	Fe–Fe	0.5	50	15	4.0	Cr(III), Cr(VI)	Cr(III), Cr(VI), 1,495, 887	100, 100	[18]
Metal holding model wastewater	Fe–Fe	0.3	250	40	3.0	Mn, Zn, Ni, Cu	Mn, Zn, Ni, Cu, 250, 250, 250, 250	72.6, 96, 96, 96	[19]
Metal coating effluent	Fe–Al, Fe–Fe, Al–Fe, Al–Al	1	100	20	7.0–9.0	Cr, Cu, Ni	Cr, Cu, Ni, 44.5, 45, 394	100, 100, 100	[20]
Metal coating effluent	Fe–Al	NA			6.0–8.0	As	As, 123	99.6	[21]
Metal coating effluent	Fe–Al	1.5	56–222	55	9.0–10.0	Cr(VI)	Cr(VI), 100	99	[22]
Metal coating effluent	Fe–Al	0.9	–	0.1–4	7.0–9.0	As	As, 20	99.9	[23]
Metal coating effluent	Al–Al	0.5	150	5–35	4.0–9.0	Zn, Mn, Cu	Zn, Mn, Cu, 50–200	100, 85, 100	[24]
Electroplating effluent	Al–Al	1.5	40	80	4.0–8.0	Zn, Ni, Cu, Cr	Zn, Ni, Cu, Cr, 300, 300, 300, 300	99, 99, 99, 99	[25]
Metal coating effluent	Al–Fe	1	100	20	9.0	Ni, Cr, Cu	Ni, Cr, Cu, 394, 44.5, 45	100, 100, 100	[26]
Metal coating effluent	Al–Fe	1	25–100	60	3.0	Ni, Cr, Cu	Ni, Cr, Cu, 526, 193, 335	99, 99.4, 98.7	[26]
Galvanic effluent	Al–Fe	2		180	5.0	Cr, Cu, Ni	Cr, Cu, Ni, 700, 500, 2,000	65, 100, 95	[27]
Automotive assembly plant rinse water	Al–Fe	1.1	60	25	3.0–6.0	Zn	Zn, PO_4^{--}, 40, 120	97, 90	[28]
Real effluent of plating industry	SS–SS	0.3	90	180	6.0–8.0	Ni, Zn, TOC	Ni, Zn, TOC, 260, 225, 173	100, 100, 66	[29]
Metal coating effluent	SS–SS	0.3	90	120	6.0	Ni, Zn, TOC	TOC, Zn, Ni, 170, 236, 282	100, 100, 66	[29]
Metal coating effluent	Fe–Fe	1.8	5	300	3.0–6.0	Cr(VI)	Cr(VI), 5	60	[30]
Electroplating effluent	Al–Fe	–	–	15	4.0–8.0	Cr(VI)	Cr(VI), 800	99.9	[31]
Metal coating effluent	Al–Fe	1.5	40	20	3.0	Cr	Cr, 500	99	[32]
Metal coating effluent	MS–MS	1.2	48.78	40	3.4	Cr(III)	Cr(III), 1,000	99.8	[33]
Industrial effluent	Al–Al	2.2	48.78	60	5.2	Cr(III)	Cr(III), 8,084	80.5	[33]
Aqueous solution	Al–Al	2.2	43.6		4.8	Cr(VI)	Cr(VI), 50	42	[34]
Metal coating effluent	Fe–Al	3.0	30	60	6.0	As	As, 1–1,230	78.9–99.6	[35]
Industrial effluent	Al–Al	1.5	40	20	1.0–7.0	Cd	Cd, 500	99	[36]
Electroplating effluent	Al–Al	5.0	48	30	4.0–8.0	Cr, Zn, Cu	Cr, Zn, Cu, 800, 800, 800	59.4, 99.9, 99.9	[37]

Table 2
Treatment of various anionic contained water/wastewater using electrocoagulation process in batch reactor

Water and wastewater used	Electrode	EG (cm)	CD (A/m^2)	Reaction time (min)	pH°	AI (mg/dm^3)	*R* (%)	References
Simulated water	MS–MS	1.5	75.4	40	3.0	Fluoride = 50 mg/L	85%	[38]
Groundwater	Al–Al	0.46	100	0.93	7.6	Arsenic = 0.22	93.2	[39]
Groundwater	Al/Al	1.2	70	0.46	7.38	Fluoride = 4.08	80.29	[40]
Well water	Fe–Al	1	–	30	7.0	Fluoride = 6.44	77%	[41]
Simulated water	SS	1.5	117	50	–	Phosphate = 500	99%	[42]
Groundwater	Fe/Al	1.5	120	40	8.74	Fluoride = 4.93	95	[43]
Groundwater	Al–Al	0.46	70	75	7.5	Arsenic = 0.23	82.17	[44]
Simulated water	Fe/air–carbon	0.3	4	8	7.0	Arsenic = 1	99.9	[45]
Simulated water	Al, air–cathode, graphite sheet	1	150	30	5.0	Phosphate = 30	30	[46]
Simulated water	Fe–Fe	1	2	180	7.0	Arsenic = 0.5	90	[47]
Simulated water	Fe–Fe	0.5	5.4	30	4.0	Arsenic = 50	99.5	[48]
Simulated water	Al–Al	0.5	5.4	60	4.0	Arsenic = 50	85	[48]
Simulated water	Al–Al	1	30 V	80	–	Fluoride = 20	60	[49]
Simulated water	Al–Al	1	37.5	30	6.0	Fluoride = 6	79	[50]
Simulated water	Fe–Fe	1	40 V	60	4.56	Cyanide = 20–50	<90	[51]
Simulated water	Fe–Al	3	15	20		Cyanide = 300	93%	[52]
Simulated water	Al–Fe, Al–Al, Fe–Fe, Fe–Al	–	–	–	–	Cyanide = 300	32, 35, 87, 93	[53]
Simulated water	Al–Al	0.5	–	–	–	Fluoride = 10	90	[54]
Simulated water	Al–Al, Fe–Fe	–	–	60	7.43	Nitrate = 105	89.7, 90	[55]
Simulated water	Al–Al	–	10	5	6–6.1	Fluoride = 5	80	[56]
Simulated water	Al/Al	–	16.7	25	7.0	Fluoride = 25	94.5	[57]
Steel industry water	Al–Al	1.5	30 V	5	–	Fluoride = 5	93	[58]
Simulated water	Fe–Al–SS	0.3	20	30	7.0	Phosphate = 100	99	[59]
Simulated water	Al–Al	1.5	30 V	80	6.0–8.0	Fluoride = 25	62	[60]
Simulated water	Al	1.5	50	50	7.0	Arsenic = 73	100	[61]
Simulated water	Fe	1.5	5	50	7	Arsenic = 73	100	[61]
Simulated water	Fe	1.5	15	55	7	Arsenic = 50	94	[62]
Simulated water	Al–Al	0.5	62.5	30		Fluoride = 4–10	90	[63]
Simulated water	Al–Al	0.5	2.2	30	3.4–7	Fluoride = 4–6	80	[64]
Simulated water	Fe–Fe	2	6	40	2.18	Fluoride = 25	40	[65]
Simulated water	Al–Al	–	–	–	–	Fluoride = 5	96	[66]
Simulated water	Al/Al	1	8.16	9	5.2	Fluoride = 25	90	[67]
Simulated water	Al–Al	0.5	0.683	30	6–8	Fluoride = 10–25	90	[68]
Simulated water	Al–Al	0.3	7.5	–	6.2	Phosphate = 10–200	90	[69]
Simulated water	Al–Al	–	–	–	–	Fluoride = 25	100	[70]
Simulated water	Al–Al	–	–	–	–	Fluoride = 16	87.5	[71]
Simulated water	Al–Al	–	–	–	–	Fluoride = 2.5	80	[72]
Simulated water	Fe–Fe	–	–	–	–	Nitrate, ammonia = 5	100, 15	[73]

Furthermore, the combination of Eqs. (4) and (5) produces aluminium hydroxide as given by the following reaction:

$$Al_3^+ + 3OH^- \rightarrow Al(OH)_3 \quad (6)$$

Hence monomeric and polymeric species forms similar to the iron electrode that settles down through the gravity.

2.3. Sweep coagulation

As earlier discussed, the formation of metal hydroxide must occur during the electrocoagulation process as a final product. Metal hydroxide is heavier than water consequently it is easily settled through gravity. Colloids may be entrapped in a flock or they may become enmeshed by its "sticky" surface as flocks settle. The removal of colloids from solution through this way is called sweep coagulation [150].

Table 3
Organics removal from different types of industrial effluent using electrocoagulation process in batch type reactor

Industrial effluent	Electrode	EG (cm)	CD (A/m^2)	Reaction time (min)	pH^o	COD^o (mg/L)	R (%)	Reference
Dairy wastewater	Al–Al	1.5	36	60	4.5	810	75	[74]
Slaughterhouse oriented wastewater	Fe–Fe	1	36	60	4.5	920	75	[74]
Automobile wash wastewater	Al–Fe	1	28	40	6.0	1,010	99	[75]
Textile effluent	Al–Al	2	25	120	5.0	1,470	18.6	[76]
Bypass wastewater	Fe–Fe	1	22	40		200	87	[77]
Laundry wastewater	Graphite-Steel	1	5.26	5	5.26	720	90	[78]
Sulfide mineral processing effluent	Al–Al	2.5	90.01	–	6.5	1,250	98.3	[79]
Palm oil mill wastewater	Al–Al	3	40.21	45.67	4.4	1,550	71.3	[80]
Oily wastewater	Al–Al	0.5	8	20	3.6	1,150	14	[81]
Biodiesel effluent	Fe–Al	0.75	32	30	6.0	404	91	[82]
Textile effluent	Al–Al	1	10–40	–	7.0	1,610	99.9	[83]
Industrial effluent	Fe–Fe	1	30	40	8.0	–	97	[84]
Palm oil mill wastewater	Al–Al	1	80	180	–	1,190	81.11	[85]
Palm oil mill wastewater	Fe–Fe	1	80	210	–	1,190	86.67	[85]
Textile effluent	Fe–Al	3	2	80	8.0	1,760	90	[86]
Urban wastewater	Al–Al	3	20	30	7.4	560	85	[87]
Biotreated municipal wastewater	Al–Al	0.5	0.265	40	7.5	330	63.21	[88]
Metalworking generated wastewater	Al–Al	1	8	25	6.5	750	25	[89]
Metalworking generated wastewater	Fe–Fe	1	8	80	7.5	750	90	[89]
Oily wastewater	Fe–Fe	2	40	40	7.0	3,500	99	[90]
Palm oil mill wastewater	Al–Al	3	56	65	4.5	1,230	75.4	[91]
Tannery effluent	Al–Al	2	400	360	6.0	580	99	[92]
Bilge wastewater	Al–Al	8	10 V	90	8.0	610	52	[93]
Bilge wastewater	Al–Al	1	10	120	7.0	21,120	85	[94]
Synthetic bilge water	Al–Al	1	10 V	120	7.0	460	85	[94]
Real bilge wastewater	Al–Al	1	10 V	120	7.0	–	89.4	[94]
Rice grain-based distillery wastewater	Cu–Cu	0.15	89.3	60	3.5	11,500	80	[95]
Paint oriented effluent	Al–Fe	1	35	15	6.95	19,700	94	[96]
Can industrial effluent	Al–Al	2	20	60		850	72	[97]
Molasses based effluent	Cu–Fe	2	33	200	8.5	4,150	54	[98]
Rice grain-based distillery wastewater	Fe–Fe	1.5–2.5	99	120	8.0	13,600	94	[99]
Rice grain-based distillery wastewater	Al–Al	1.5	89.3	60	8.0	13,800	93	[100]
Oil tanning industrial wastewater	Al–Fe	1.5	40	10	6.74	21,000–25,000	94.44	[101]
Pulp and paper mill effluent	Al–Fe	1	70	60	5.0–7.0	1,700	87	[102]
Textile industrial wastewater	Al–Fe	–	–	–	–	1,953	93	[103]
Slaughterhouse based wastewater	Al–Fe	–	–	–	–	4,000	96.6	[104]
Tannery liming drum effluent	MS–Al	2.5	35	50	3.0	25,300	82	[105]

(Continued)

Table 3

Industrial effluent	Electrode	EG (cm)	CD (A/m^2)	Reaction time (min)	pH^o	COD^o (mg/L)	R (%)	Reference
Industrial wastewater	Fe–Al	–	45.45	60	8.0	2,000–2,500	69	[106]
Petroleum refinery effluent	Al–Fe–SS	–	13	60	8.0	4,090	63–93	[107]
Tannery effluent	MS–Al	1	–	60	8.5	2,400–2,600	68	[108]
Rose processing wastewater	Fe–Fe	–	–	–	–	9,500	79.8	[109]
Molasses based distillery wastewater	Fe–Fe	–	60	–	4.0	–	92.6	[110]
Food processing wastewater	Al–Al	1	–	–	–	22,956	88	[111]
Refectory oily effluent	Fe–Fe	1	–	100–150	–	500–1,500	75	[112]
Chemical mechanical polishing wastewater	Al–Fe–Ti	–	–	–	6.0–8.7	500	75	[113]

2.4. Interparticle bridging

When the metal anode is dissociated in water, large molecules may also form, or may be synthetic polymer occurs. These polymers maybe linear or branched and their surface may be highly reactive. Consequently, available colloids of the solution may combine to one polymer or several and settable mass is the final outcome [151].

3. Types of electrocoagulation

The electrocoagulation process usually performs in two ways namely (1) batch process and (2) continuous process. A brief description of both processes is given below:

3.1. Batch electrocoagulation process

In a typical batch electrocoagulation process, the retention time of colloids was uniform throughout the process. A fixed amount of water/wastewater is filled in a batch reactor equipped with metal electrodes. This whole arrangement is stirred by a stirrer. The pH of the solution is either fixed before the reaction or maintain throughout the process using diluted acid or base. The current density and electrode gap are maintained uniform within the entire process. In the fixed time interval, the small quantity of treated water is taken out from the reactor and various parameters are determined. In other words, batch electrocoagulation is steady-state throughout the process with constant volume, fixed reaction time, and initial pH, however the generation of metal ions and their dissolution varies depending upon the number of residual organics in bulk solution [52].

3.2. Continuous electrocoagulation process

In a continuous process, influent continuous supplies in a reactor with a fixed flow rate. In other words, the input and output rate of the reactor is equal during the whole process. The reactor is continuously stirred using a stirrer. The current density is maintained uniform throughout the process. The discharge received from the reactor is analyzed for the measurement of various parameters. The requirement of constant *in-situ* coagulant is provided by the efficient design and operational control. The continuous process is efficient as compared to batch process in the same cost however the quality of treated effluent is poor in continuous process as compared to batch process output [152].

4. Operating parameters

The performance of electrocoagulation depends on the operating parameter of the process. In other words, at the optimum condition of operating parameters, this process can achieve better results. There are several operating parameters of the electrocoagulation process, however, pH of solution, current density, electrode gap, electrolysis time, and anode material are the main influencing parameters [153]. The detailed description of the main influencing parameters and other influencing parameters are as follows:

4.1. Selection of electrode materials

The selection of electrodes for the electrocoagulation process has immense importance because the separation efficiency of pollutants from water/wastewater depends on the nature of metallic coagulant that generates during the process. For example, removal of fluoride from the solution is well achieved through aluminum electrode instead of the iron electrode. In other words, selection of electrodes is depended on the nature of various pollutants/contaminants of the wastewater [154].

Usually, iron and aluminium are used as the sacrificial electrodes for the electrocoagulation process to treat different types of wastewater. Cast iron and stainless steel are other useful materials that can also be considered as an electrode. Apart from this, some special types of electrodes such as BDD (Boron-doped diamond), nickel, graphite, PbO_2, and SnO_2 has also been practiced in rare electrocoagulation processes. However, special type electrodes have dimensionally stable anodes (DSA) but these types of materials are used in some specific effluent, also it can enhance the cost of the process. For example, BDD, SnO_2, PbO_2, nickel, and graphite electrodes show greater

Table 4
Treatment of different water/wastewater using electrocoagulation process combination with other methods

Industrial effluent	Grouping	Outcomes (%)	Initial pollutants concentration (mg/L)	References
Distillery effluent	Electrocoagulation and coagulation	100%	9,080	[114]
Electroplating effluent	Persulfate enhanced electrochemical oxidation	COD = 95.8, TOC = 87.8 and CN = 98.4	COD = 11,290, TOC = 4,456 and CN = 1,280.15	[115]
Electroplating effluent	Electrooxidation with coagulation–flocculation	100	COD = 233	[116]
Textile effluent	Electrocoagulation with adsorption	100	COD = 18,600	[117]
Industrial effluent	Electrocoagulation with floatation	Nitrate removal of 70%	300	[118]
Electroplating effluent	*In-situ* ion-exchange electrocatalysis biological coupling	COD = 87.23, TOC = 80.42, Cr = 91.25, Cu = 95.97	COD = 274.89–319.42, TOC = 89.55–96.40, Cu^{2+} = 0.056–0.137, Cr^{6+} = 0.442–1.111	[119]
Real wastewater	Electrocoagulation coupled with adsorption	TOC removal of 89%	500	[120]
Electroplating effluent	Electrooxidation–electrodeposition	Ni = 99, NH_3 = 70	Ni = 2,156, NH_3 = 9,680 mg/L	[121]
Fabric manufacturing effluent	EC, electrochemical Fenton (ECF), electro-Fenton (ECF) and peroxi-coagulation (PC)	COD and color removal of 78.6% and 77%–94% respectively using EC COD reduction of 82.1%, 64.2% and 71.1% using ECF, EF, PC respectively	COD = 1,310	[122]
Electroplating effluent	Reduction/precipitation, chemical oxidation and biological aerated filter	Cu = 9.94, Cr = 99.5, Ni = 99.0, CN = 99.7, COD = 84.2	Cu = 108, Cr = 62, Ni = 85, CN = 136, COD = 450	[123]
Winery effluent	Ozone supported electrocoagulation	COD = 83	COD = 2,500	[124]
Simulated wastewater	Advanced oxidation process followed by electrocoagulation	Cr = 99	–	[125]
Industrial effluent	Electrocoagulation combined with reverse osmosis membrane	Turbidity = 135, COD = 280	Turbidity removal of 93.80% and COD removal of 66.64%	[126]
Industrial effluent wastewater	Electrocoagulation coupled with chemical coagulation	High removal percentage of arsenic and remaining is reduced by chemical coagulation	–	[127]
Electroplating effluent	Electrocoagulation–electrooxidation	COD < 99 and BOD < 99	–	[128]
Laundry wastewater	Electrocoagulation followed by electroflotation	999 mg/dm^3 COD removal of per kWh of energy consumption	–	[129]

potential against chemical resistance and are well capable to degrade cyanide from the wastewaters. Another example is PbO_2 and graphite not easily dissolve in water while that used as anodic material and commonly put in electroflotation (EF) process [155,156]. These anodic materials are less expensive and are commonly available. However, during the EC process a highly toxic Pb^+ is generated by PbO_2 anodes that lead to serious secondary pollution. Furthermore, Ti/PbO_2 or Ti/SiO_2 anodes provided similar results compared to IrOx, Ti/IrOx-Ta_2O_5 electrodes while Ir oriented electrodes are more expensive. Also, Ti/PbO_2 or Ti/SiO_2 anodes have a better ability to oxidize toxic compounds [157]. If the solution contains Mg^{2+} and Ca^{2+} ions in significant amounts, SS electrodes can be used [158].

Table 5
Treatment efficiency of electrocoagulation process as compared with other treatment methods

Industrial effluent	Comparison	Outcome	Concentration of effluent (mg/L)	References
Distillery effluent	Electrocoagulation and chemical coagulation	EC = 91%, chemical coagulation = 85%	9,080	[130]
Distillery effluent	Electrocoagulation and sono-electrocoagulation	EC = 85%, sono-electrocoagulation = 99%	8,000	[130]
Industrial effluent	Electrocoagulation and electroflotation method	92% COD reduction using EC coupled with EF, 97% of COD reduction using individual EC	2,983	[131]
Gelatin production wastewater	Electrocoagulation and chemical coagulation	73.6% COD reduction using EC and 55.6% COD reduction using coagulation	1,560	[132]
Simulated purified terephthalic acid wastewater	Electrocoagulation, peroxi-electrocoagulation (PEC) and peroxi-coagulation (PC) processes	EC = 60.76, PEC = 73.91, PC = 66.68%	75	[133]
Simulated solution	Anodic oxidation (AO) and electrocoagulation (EC)	87% dye removal by EC and 75% dye removal by AO	100 mg/dm^3	[134]
Textile factory	Electrocoagulation and chemical coagulation	EC: 58.86%, coagulation = 56.08%		[135]
Tannery effluent	Hybrid electrocoagulation (HEC) and electrodialysis process (EDP)	73% of COD reduction by HEC and 92% of COD reduction using HEC + EDO	2,800	[136])
Municipal wastewater	Electrocoagulation, biological treatment	EC = 84, biological treatment = 83	890	[137])
Electroplating effluent	Electrocoagulation and chemical coagulation	Complete removal of Cr(III) and Cr(VI) using EC and 52.6%, 25.8 Cr(III) and Cr(VI) removal respect using chemical coagulation	Chromium(VI) 887 Chromium(III) 1,495	[138]
Plug board manufactured effluent	Electrocoagulation (EC) and electrocoagulation + H_2O_2	EC: 30%, EC + H_2O_2: 76%	COD = 242	[139]
Simulated dye solution	Electrocoagulation and chemical coagulation	98% of COD reduction using EC and 53% of COD reduction using chemical coagulation	200	[140]
Textile wastewater	Electrocoagulation and chemical coagulation	98% of COD reduction using EC and 53% of COD reduction using chemical coagulation	1,890	[141]
Aqueous suspension of kaolinite	Electrocoagulation and chemical coagulation	99.9% of COD reduction using chemical coagulation and 96% of COD reduction using EC	200	[142]
Palm oil mill effluent (POME)	Electrocoagulation and chemical coagulation	EC: COD = 68%, coagulation = 55%	–	[143]
Textile effluent	Electrocoagulation and chemical coagulation	EC: 23%, EC + alum: 65%, EC + poly aluminum chloride: 80%	COD = 3,422	[144]

4.2. pH of solution

The initial value of pH of any solution (pH_i) has played an important role in the efficiency of the ECT. The removal of pollutants from wastewater depend on the balance among H^+ ions, the amount of coagulants generates during electrocoagulation process, and negative ions produced by the colloids [159,160]. In other words, the role of pH is mainly related to the discharge of hydroxide ions by coagulant during the removal of pollutants using electrocoagulation. However, with higher electrolysis time the changes in pH are meaningless because the solution tries to shift to neutral pH. Additionally, higher current density promotes the formation of water molecules consequently pH rises with this condition is least meaningful [161].

$$H^+ + OH^- \rightarrow H_2O \quad (7)$$

The optimum pH is varies for different types of wastewater. For example, in the case of molasses-based distillery wastewater optimum pH is varied in the range of 4–5

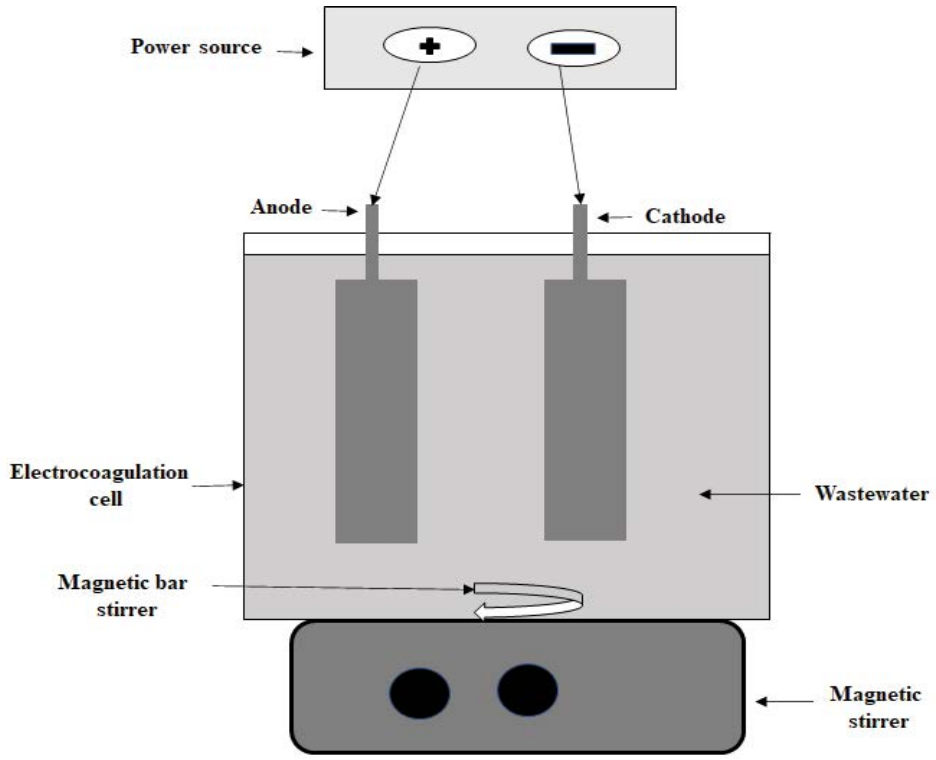

Fig. 1. Experimental setup of electrocoagulation process.

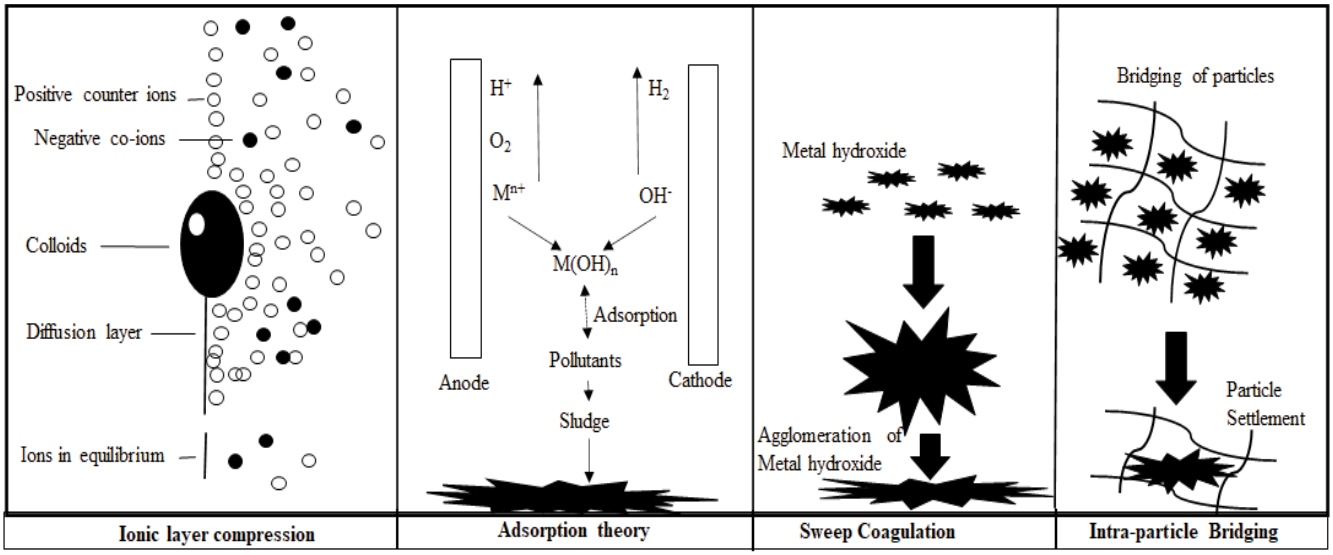

Fig. 2. Different mechanisms of electrocoagulation process.

[162]. In the case of the metal plating industry, optimum pH varies in the range 3.5–7 while rice grain-based distillery wastewater provided better organic reduction at pH range of 7–8.5 [10,99]. However, this condition is not always true because distillery, metal plating effluent, textile industry do not always contains the common type of pollutants [163]. The presence of pollutants also depends on raw material, chemicals, reactor, and process used during the process.

4.3. Current density

Current density is a main game-changer of the electrocoagulation process, usually defined as the ratio of the current across the electrodes divided by the active area of the electrode (A/m^2 or mA/m^2). The removal of pollutants is mostly subjected to the number of metal ions generated during the electrocoagulation process. The rate of metal ions

production is directly propositional to the current density as given by Faraday's law [164];

$$m = \frac{It_s M}{ZF} \tag{8}$$

where m represents the amount of metal ions; F is known as Faraday's constant; Z represents the valency of the ions m; I is the value of applied current (A); t_s is the reaction time in minutes, and M represents the molar mass of electrode material. Sufficient availability of metal ions enhances the removal rate of pollutants; however, overdosing of metal ions can restabilize the particles [165]. Moreover, with an increase in the current density bubbles generation rate increases, which in turn increase the separation effect. The bubbles size increases with a decrease in the current density. Both are beneficial for high pollutant removal efficiency due to the formation of H_2 [166].

For design purpose the optimization of current density is very important because the electrode and energy consumption increases with an increase in CD, also, a stronger current (higher current density) results in loose flocs which settle slowly as they contain a large amount of bound water and produces a clearer supernatant [167].

4.4. Electrode gap

Inter electrode gap also influences the performance of the electrocoagulation process [121,168]. For the maximum removal efficiency of pollutants, the electrode gap should be optimum. The larger electrode gap enhances the cost of the process because it increases resistance offered by the cell as presented by following Faraday's equation as [159].

$$R = \frac{g}{KA} \tag{9}$$

where K represents the cell specific conductance of solution, and A represents electrode surface area. In other words, higher electrode gap decreases both local concentration and electrostatic attraction consequently removal of pollutants decreases. Lesser electrode gap reduces the cost of the process but it avoids proper circulation of solution consequently influencing the process efficiency [5].

Chen et al. [169] reported ohmic potential drop increases with an increase in inter-electrode spacing. Hence, to reduce the cost of electrical energy, electrode space should be minimal. Narrower gaps are responsible to enhance mass transfer coefficient, and also increase ohmic loss. Decrease in gap may increase the electrolyte resistance (i.e., when the electrode gap is narrower no need of electrolyte). We can reduce electrode gap minimum of 8 and 3 mm when handling real solution and synthetic solution, respectively. However, a narrow gap of less than 1 cm is conveyed with small electrical energy consumption. Also, with greater electrode gap, cell voltage enhances [170] consequently power consumption of the system increases. When planning for high SA/V reactors, a sufficient electrode gap is required for sufficient turbulence of solution in-between the electrodes that help to initiate mass transfer within the electrocoagulation reactor and improve the removal efficiency of the process. If the electrode gap is less than 10 mm, it can reduce the swirling velocity of the solution between the electrodes thus removal efficiency of pollutants affected [171].

4.5. Electrolysis time

In the design point of view, electrolysis time is very important during the electrocoagulation process because from Faraday's law [Eq. (8)] that the amount of metal ions discharges is directly proportional to the electrolysis time consequently pollutant reduction efficiency enhance with an increase in reaction time [172]. However, at higher current density the need for electrolysis time is less because, with an increase in current, anodic dissolution rate also enhances. In addition, with increase in ionic concentration in solution is generally responsible for an increase in current intensity at the same cell voltage [114]. In addition, as a balanced amount of metal ions is required for the maximum removal of pollutants hence overtime of reaction can destabilize the particle consequently pollutant removal efficiency decreases [173].

4.6. Electrode thickness

The electrode thickness should be optimized in the electrocoagulation process. If the thickness of an electrode is more, it can reduce the value of actual current passed while the voltage of the system unnecessarily increases consequently increasing the cost of the process [174,175]. However, wider and thicker electrodes could minimize drop of potential and also have long life, and are easily maintained during backwash operations of the electrocoagulation reactor. Most of the researcher works on electrode thickness around 2 mm. However, it also depends on electrode material. For example, aluminium electrode is thicker than SS-302 electrode even for the same solution [176].

4.7. Cell potential

During the electrocoagulation process, cell voltage appears due to the potential difference occurs between two identical metal used for both cathode and anode. When current is supplied in electrode, the cell voltage comprises of several components, such as the potential drops due to the bulk resistivity of the electrolyte and the potential drop due to the resistivity of the leads, the potential differences associated with the electrochemical processes occurring on the cathode and the anode [177]. Undesired cell potential can reduce the efficiency of electrocoagulation reactor hence its optimization is required.

4.8. Limiting current

When the potential occurs between the anode and cathode the value of the limiting current is achieved. This parameter plays very important role to control the mass transfer coefficient of the system and can be expressed as [178];

$$K_m = \frac{I_{\text{Lim}}}{n\text{FAC}_b} \tag{10}$$

where I_{Lim} is limiting current, A represents anodic surface area (m^2), n represents the amount of exchanged electrons in electro-oxidation reaction, K_m represents the value of mass-transfer coefficient (m/s), F represents Faraday constant (96487 C/mol) and C_b represents the concentration of organics in solution (mol/m). In other words, for higher concentration of organics, limiting current should be higher for the better efficiency of EC process [179].

4.9. Bubble size distribution and its control

Eqs. (2) and (5) indicate hydrogen grown in the cathode surface while at the anode oxygen is evolved. In other words, due to the presence of gases at the electrode surface which promotes bubbles production and in the gap of the electrode, it works as an insulator [180]. This condition is not favorable for electrocoagulation process and may reduce its efficiency. In addition, as bubbles work as insulator, it can enhance the electrical resistance. This problem could be overcome, to introduce electrolyte flow between two electrodes that induce turbulence and sweep out the gas bubbles [181]. However, bubble size distribution mainly depends on electrode material and pH of the solution [182].

4.10. Electrode passivation and flake deposition

When electrodes are used for long period, a scale of magnesium carbonate or calcium carbonate is existed at surface of electrode [182]. However, scale formation may occur in short period also, depends on the quantity of calcium/magnesium present in the water. Flake deposition on anode also is an important issue that influences the efficiency of electrocoagulation process [183]. Overall, both electrode passivation and flake deposition enhance the electrical resistivity and reduces metal ions production rate consequently the reduction efficiency of pollutant during electrocoagulation process decreases [184]. To avoid this type problem a time interval should be fixed for the replacement and cleaning of electrodes are needed.

4.11. Polarity reversal

Metal ions are releases from the anode surface causes weight loss of anode while cathode gains some weight. In addition, due to the flake deposition on the anodes offers high power consumption and least pollutant removal efficiency [184]. To overcome this problem, Lach et al. [185] performed a series of electrocoagulation experiments to examine the way of flake deposition, and its cleaning by reversing the polarity of the electrodes.

4.12. Temperature

Usually the rate of electrocoagulation reactions enhances with rise in temperature and could be expressed by following Arrhenius expression [186];

$$k = k_0 \exp\left(\frac{-E_a}{RT}\right) \tag{11}$$

where k represents the constant of the reaction rate (the unit of k is depends on the order of the reaction), E represents activation energy of the process (kJ/mol), k_0 represents he frequency factor having unit is same of k, T represents absolute temperature of the system (K), R represents gas constant (8.31541 J/(mol K)). However, most of electrocoagulation processes have been performed in atmospheric condition. Furthermore, temperature is little influences the results of electrocoagulation process. Ribordy et al. [187] reported that that with variation in temperature from 20°C to 60°C, electrochemical degradation of flavor manufacturing effluent did not while Platinum is used as anodic material.

4.13. Conductivity

For the run of electrochemical process, a minimum level of conductivity should be present in solution. In some effluent conductivity level is good hence it need not to be maintained. However, effluent that suffering lake of conductivity calculated amount of electrolytes such as NACl PAA etc. should be added in effluent [188].

The conductivity of solution directly influences the need of energy required for accomplish a desired level of performance during electrocoagulation process [189]. Hence to minimize the electrical energy consumption the conductivity of solution should be sufficient.

4.14. Effect of agitation speed

During the process, a defined agitation speed is required to uniform mixing of solution and also protects the production of the concentration gradient. Moreover, velocity of generated ions is managed through agitation of solution. For the maximum removal of pollutants from the wastewater agitation speed should be optimum [190]. The overspeed of agitation can disturb the system of reactor because flooding of solution and degradation of flocks inside the reactor. However, poor speed of agitation influences the contact of metallic ions and negative ions discharges by colloids [191].

4.15. Shape of electrode

The amount of metal ions production is oriented on the effective surface area (i.e. area that actually is in contact of solution) of the electrode [191]. For example, if electrode is circular then metal ion production is less while energy consumption is high as compared to the rectangular electrode [192]. However, the shape of electrodes also determined on the basis of size and type of reactor used in experiments. Perforated electrodes release larger amount of metal ions as compared to the plane electrodes. As per open literature few publications have been reported about the effect of electrode shape on electrocoagulation process [81]. These researchers found that plain electrode discharges lesser amount of metal ions as compared to perforated electrode. In the case of perforated electrode, the intensity of the electric field at the corner edge was (1.2) times higher than the plain electrodes consequently increase in output current in the perforated type electrode.

4.16. Arrangement of electrode

In the cost optimization point of view both the type of electrode material and the mode of connection have

immense important. Both monopolar and bipolar electrode connections are used for electrocoagulation process. In bipolar arrangement more than a pair electrode fitted in the reactor and only the electrodes are connected in respective anode and cathode through DC supplier. Bipolar connection provided higher surface area as compared to monopolar connection due to two electrochemical cells acted together and favored the adequate anodic oxidation [67]. Hence pollutant removal efficiency of bipolar electrode is higher than monopolar connection because the intensity of current is higher.

4.17. Cost optimization

In the cost point of view electrical energy consumption is an important parameter during the electrocoagulation process. Therefore, its calculation is made by equation below,

$$P\left(\frac{\text{Wh}}{\text{dm}^3}\right)=\frac{VIt}{\text{Treated volume}} \tag{12}$$

where V stands for the cell voltage in volts, I stands for the current in amperes, and t stands for time in hours. It can be seen from Eq. (12) that energy consumption increases with increase in current and time hence during the EC process the value of I and t should be optimum.

5. Future scope

This article elucidates that electrocoagulation is a promising method to for the removal of pollutants from various water/wastewaters. For the commercial implementation of electrocoagulation technique, the laboratory scale batch experiments are required. Also, knowledge obtained from the batch process should be also applied to continuous process. This review article can help to future research using following recommendations:

- Electrocoagulation method has a great potential to treat other pollutants such as containing chlorides, sulfates organics and dyes.
- Most of electrocoagulation method accomplished by using either iron or aluminium electrode. However, other electrode such as graphite, SS can also be implemented to treat industrial wastewater using either series or bipolar arrangement. Also, research should be extended for both batch and continuous electrocoagulation reactors and on the basis of their beneficial comparison adoption should be made.
- Since the electrocoagulation in single step are unable to remove the organic load completely, it is essential to further treat the wastewater by other post-treatment options such as adsorption, membrane separation to achieve the required discharge standards of the effluents.
- Electrocoagulation followed by the other physicochemical treatment method may be used for industrial scale to meet the stipulated regulatory discharge standards of the effluents.
- During electrocoagulation process sufficient amount of hydrogen gas is generated, the gas composition needs to be analyzed.
- The adsorptive characteristics of the electrocoagulation generated sludge should be studied.

6. Conclusions

From this review article following conclusion could be made:

- Electrocoagulation process is better choice to treat not only electroplating effluent but also it can be used for treatment of different type of wastewater.
- Electrocoagulation is based *in-situ* production of metal ions which added with negative ions passes by different functional groups of organics or colloids, hence no need to addition of chemical coagulant.
- This method has less installation cost and also required less maintenance.
- As compared to other treatment process such as coagulation/flocculation, adsorption, membrane separation and wet air oxidation, electrocoagulation is more acceptable. Because during coagulation large amounts of sludge are generated that enhances maintenance cost. Regeneration of adsorbent is the limitation of adsorption process. Less productivity and high cost are major disadvantages of membrane process while wet oxidation process required special operating condition in term of high temperature and pressure.

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Declaration of competing interest

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