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A Review of Electrocoagulation Process for Wastewater Treatment

Bharath M*, Krishna B M and Manoj Kumar B

[1]Department of Environmental Engineering, Sri Jayachamarajendra College of Engineering, Mysuru – 570006, Karnataka, India.

Abstract : The control of environmental pollution and also the treatment of polluted water are of great concern. Within the past decade, electrochemical coagulation process has emerged as most effective wastewater treatment process as compared to conventional techniques of treating wastewater. Electrocoagulation is robust, cost effective, reliable, low sludge generating process, it has automation amenability and it has high pollutant removal efficiency. It has been proved effective in treating various types of wastewater but is seldom accepted. The aim of the review is to explain the basics and up to date advancement of electrocoagulation method for the improvements in the pollutant removal efficiency. In this review paper, an overview of electrocoagulation method with effect of key operational parameters on it is provided. Limitations of the method are also represented for the better understanding of the mechanism of pollutant removal and its optimization. The recent advancements and future scope of the electrocoagulation process are also reviewed.

Keywords : Electrocoagulation; wastewater; poly hydroxides; sacrificial electrode.

1. Background of Ec Treatment

Electrolysis is a method in which oxidation and reduction occur due to application of electric current to the electrolytic solution. Electrochemical technology has shown to be a hopeful technique for the destruction of organic pollutants in the wide collection of wastewater and there is no need for adding additional chemicals. In addition, the high property of the electrochemical process prevents the assembly of unwanted by-products. And it can also be used for the metal recovery from the different wastewater.

In the 19th century (1889) in London, the electrochemical method was proposed with a well-established plant for the sewage treatment. In this process, wastewater was electrolyzed by mixing with sea water. The prime interest of primary stage development of the EC process was to generate chlorine for the removal of odor and disinfection of sewage wastewater. Electrochemical processes include: electro-coagulation, electro flotation, electro oxidation, electro-flocculation, electro-disinfection, electro reduction, electro-deposition, etc. Electrocoagulation (EC) is the most established electrochemical process. EC process was developed and patented by A. E. Dietrich in 1906 for the treatment of blige water from ships. Later in 1909 in the US, wastewater treatment by the electrocoagulation using aluminum and iron electrodes was proprietary by J.T. Harries. In 1984 in the US for the first time, a large scale drinking water treatment by electrocoagulation method was implemented.

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Electrocoagulation (EC) finds its application in treating different types of process wastewater, for example electroplating wastewater[1], Heavy oil refinery [2], textile wastewater[3], [4], Dairy wastewater[5], Distillery wastewater[6], [7], Leachate wastewater[8], [9].In recent years EC process has successfully applied for the de-fluoridation of groundwater[10].

In the 20th century electrochemical process had narrow success and acceptance in spite of being a competitive and effective treatment technique for most of the wastewater. The initial improvements were in minimization of electrical power consumption and throughput rates of effluent. Therefore present study focuses on the mechanism of EC process and operational factors (voltage, current density, temperature, time of treatment, electrode arrangement, inter electrode distance and pH) effecting its efficiency for the improvement of the EC process.

1.1 Electrocoagulation

Electrocoagulation process involves oxidation and reduction reaction in which destabilization of contaminants (suspended, emulsified, or dissolved) happens because of application of electric current to the electrolytic solution. EC unit consists of an electrolytic cell and metal (Al or Fe) electrodes which are connected to an external power supply. The conductive metal plates are well known as ‘sacrificial electrodes’ which are made up of same or completely different materials as anode or cathode. In the EC process, anodic dissolution generates *in situ* coagulants along with hydroxyl ions and hydrogen gas at the cathode. These *in situ* coagulants cause the formation of flocs within the sort of metal (Al or Fe) hydroxides and/or poly hydroxides. The hydrogen gas generated at the cathode brings flocs at the water surface by providing further buoyancy. The benefits and drawbacks of EC process are given below.

1.1.1 Benefits of Electrocoagulation Process

1. EC involves artless equipment and is easy to work.
2. EC requires low investment, maintenance, energy, and treatment costs.
3. EC treated wastewater furnish pleasant, odorless, clear and colorless water.
4. EC is a low sludge producing process. EC generated Sludge is mainly composed of metallic oxides/hydroxides.
5. There are no additional chemicals required in EC process.
6. Flocs formed by EC are similar to chemical floc. EC flocs are much larger in size, enclose less bound water and are acid-resistant and more firm.
7. The reuse of EC produced effluent contributes to a lesser water recovery cost because it contains a lesser amount of total dissolved solids (TDS) as related with chemical treatments.
8. The gas bubbles generated at the time of electrolysis can proceeds the pollutants to the top of solution from where it can be separated without difficulty.
9. EC provides greater efficient pH range and pH neutralization result and can be suitably used with other renewable sources of energy.

1.1.2.Disadvantages of Electrocoagulation Process

1. The sacrificial anodes are dissolved into solution due to oxidation, and need to be replaced at regular interval.
2. Conductivity of the wastewater suspension must be high.
3. Viscous hydroxide may be likely to solubilize in some cases.
4. The electricity may be not easily available and expensive in some area.
5. The efficiency of the electro coagulation unit decreases due to an impervious oxide film shaped on the cathode.

1.2. Mechanism of Electrocoagulation

The EC reactor configuration varies with the number of electrodes and arrangement of electrodes (monopolar or bipolar).

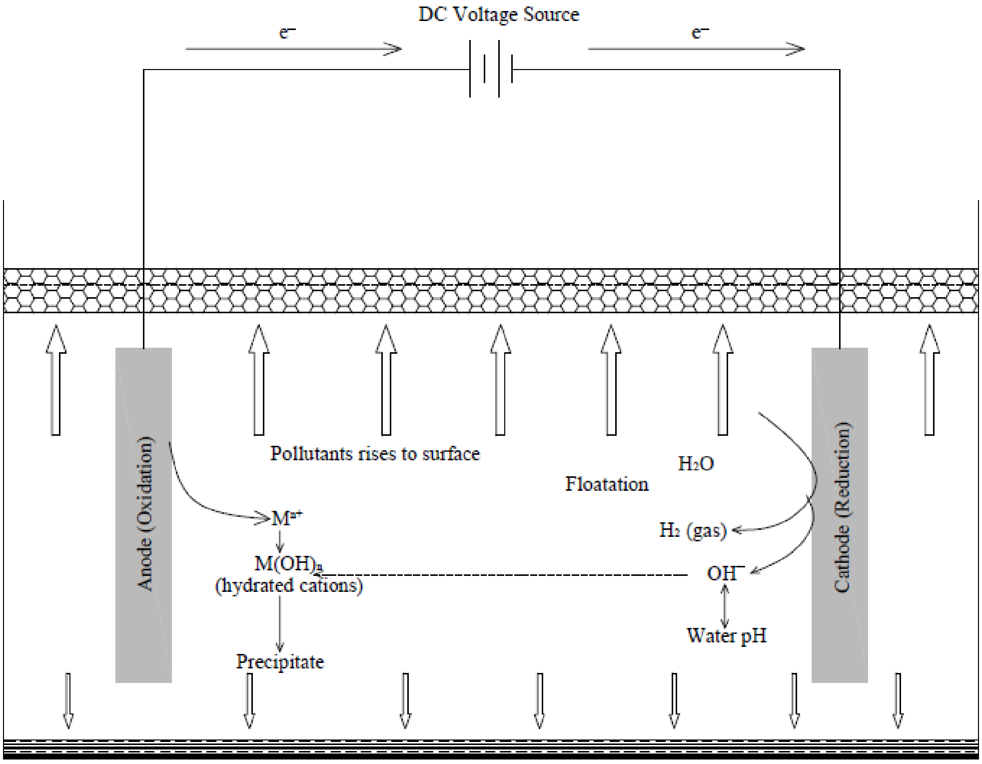

Figure 1. Diagram of a bench-scale EC reactor

The reaction mechanism of the electrochemical method using aluminum and iron electrodes is shown in Figure 1. On an applied electric current, oxidation of anodic material and reduction of cathodic material takes place.

Anodic reactions:

$Al(s) \longrightarrow Al^{3+} + 3e^-$
$Fe(s) \longrightarrow Fe^{2+} + 2e^-$
$2H_2O\,(l) \longrightarrow O_2\,(g) + 4H^+ + 4e^-$
Anode oxidation or atmospheric oxygen oxidizes ferrous ion to Fe^{3+}.
$Fe^{2+} \longrightarrow Fe^{3+} + e^-$
$2Fe^{2+} + 1/2\, O_2\,(g) + H_2O\,(l) \longrightarrow 2\, Fe^{3+}$
Cathodic reactions :
$2H_2O + 2e^- \longrightarrow H_2\,(g) + 2OH^-$
Moreover, in the presence of chloride and high anode potential, the subsequent reactions may occur in the EC cell:
$2Cl^- + Cl_2 + \longrightarrow 2e^-$
$Cl_2 + H_2O \longrightarrow HOCI + CI^- + H^+$
$HOCI \longrightarrow OCI^- + H^+$

Electrochemically produced metal cations (Fe^{3+} or Al^{3+}) will react spontaneously and produce corresponding hydroxide and /or poly hydroxides according to complex precipitation kinetics. Many monomeric species of Al^{3+}such as $Al(OH)^{2+}$, $Al(OH)^{2+}$, and $Al(OH)^{4-}$ and polymeric species such as $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}\,O_4(OH)_{24}^{4+}$, and $Al_{13}(OH)_{34}^{4+}$ transform into $Al(OH)_3$.Similarly Ferric ions species such as $FeOH^{2+}$, $Fe(OH)_2^{+}$, $Fe_2(OH)_2^{4+}$,$Fe(OH)_4^-$,$Fe(H_2O)_5\,OH^{2+}$, $Fe(H_2O)_4(OH)_2^{+}$,$Fe(H_2O)_8(OH)_2^{4+}$and $Fe(H_2O)_6(OH)_4^{2+}$ transform in to $Fe(OH)_3$. These insoluble $Fe(OH)_3$ and $Al(OH)_3$ remain in the aqueous medium and destabilize contaminants by completion or electrostatic attraction followed by coagulation. The formation of these complexes is highly pH dependent. Destabilization of contaminants mainly occurs through two distinct mechanisms, i.e.

(i) Cationic hydrolysis products neutralize negatively charged colloids.

(ii) Sweep flocculation: Entrapment and removal of contaminants in the form amorphous hydroxide precipitate.

Gas bubbles H_2 and O_2 adhere to agglomerates which are released at the electrode surfaces during electrolysis and carry them to the water surface [11].

2. Effects of Operational Parameters on EC Process

To achieve the maximum removal efficiency using electrocoagulation process in minimum electrolysis time with minimum operational cost, it is essential to understand the effect of various operational parameters on EC process.

2.1. Electrode Arrangement

In the EC process, electrode material and type of electrode connection play a major role in the cost analysis.

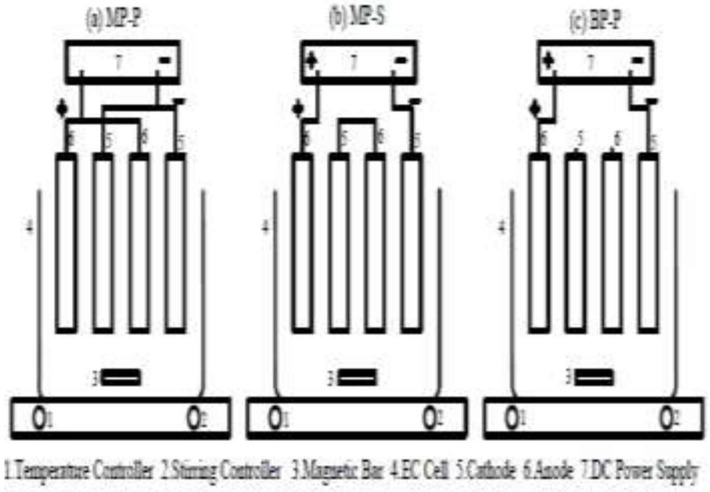

Figure 2. Diagrammatic representation of different mode of electrode connection (a) monopolar parallel (b) monopolar series and (c) bipolar parallel

The diagrammatic representation of different types of electrode connection is shown in Figure 2 (a), (b) and (c).

In the EC process, electrode materials define the type of electrochemical reaction that will occur in the EC processes. Al or Fe plate can be used as the anode and inert material such as steel, stainless steel, platinum coated titanium etc. can be used as cathode [12]. In few cases, similar material is used for the anode and cathode. An EC system can include either one or multiple anode-cathode pairs and may be connected in either a monopolar or a bipolar mode.

Monopolar mode of parallel connection (MP-P) is shown in Figure 2 (a). During this connection, current is split between all the electrodes in regards to the resistance of individual electrodes. A low potential difference is needed as compared with serial connections. In the monopolar mode of series connections (MP-S), each pair of sacrificial electrode is internally connected with one another as shown in Figure 2(b).

In the bipolar mode of parallel connection (BP-P) as shown in Figure 2(c), sacrificial electrodes are placed between the two parallel electrodes without any electrical connection. Bipolar electrode arrangement has simple set-up and hence the maintenance is low. Once an electric current is passed through the outer electrodes, the uncharged sites of conductive plates get charged with the opposite charge compared to the parallel side beside it. In the EC process, positive sides of electrodes undergo anodic reactions and negative side undergoes cathodic reactions throughout electrolysis [13].

2.2 Effect of Electrolysis Time

Electrolysis time also has significant effect on pollutant removal efficiency of electrochemical coagulation method. It defines the amount of coagulant formed and cost of the process. An increase in electrolysis time up to the optimum level increases the pollutant removal efficiency but it does not increase beyond optimum level. The actual fact is that at constant current density coagulant formation increases with an increase in electrolysis time which leads to increased removal efficiency. Whereas the above optimum electrolysis time and increase in coagulant dose does not increase the pollutant removal due to the presence of sufficient number of flocs[14]. Electrolysis time has a negative impact on cost of treatment due to increase in energy and electrode consumption at longer electrolysis time.

2.3 Inter Electrode Distance

In the EC process, inter-electrode distance plays an important role on EC potency because the electrostatic field depends on the distance between the anode and the cathode. An optimum distance between electrodes provides maximum pollutant removal efficiency. Minimum inter-electrode distance provides low pollutant removal efficiency. The more the inter electrode distance the slower the movement of the generated ions. Due to the slower movement ions gets extra time to form floc required for the coagulation of pollutants[14].Whereas an additional increase in inter electrode distance above optimal value decreases anodic dissolution and will increase the distance that ions essential to travel for floc formation, which results in the decrease in the electrocoagulation efficiency [15].

Electrocoagulation efficiency depends upon conductivity of the solution. As shown in the equation below the electrical conductivity is directly proportional to the inter electrode distance. An increase in distance between the anode and the cathode (g), increases resistance (R) offered by the cell [16].

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

Where K is the cell specific conductance and A is electrode surface area. According to Ohm's law current increases with the decrease in resistance. An increase in current leads to an increase in anodic dissolution thus increasing the electrocoagulation efficiency.

2.4 Effect of Current Density

One of the most significant operational parameters in electrocoagulation process is current density i.e., current per area of the electrode. According to the literature a wide range of current densities applied between 1-100 mA/cm^{2}depending on the case study. The separation processes which involves flotation cells or large settling tanks requires high current density, whereas sand and coal filter integrated EC process needs low current density.

The amount of electrode dissolution is directly proportional to the amount of current passed through the electrolytic solution. Faraday's law explains the relationship between current density (mA/cm^2) and the amount of metal (m) dissolved (g of M/cm^2).

$$w = J \times t \times \frac{m}{n} \times F(4)$$

Where the quantity of electrode material dissolved (w) in g /cm^2; applied current density (J) in mA/cm^2;electrolysis time (t) in s; m the relative molar mass of the electrode material under study; n the number of electrons in oxidation/reduction reaction; and F is Faraday's constant (96,500 C/mol).

At high current density, there is wastage of electrical energy in heating up of water which results in decrease in current efficiency. To ensure a high current efficiency, the current density must be selected in accordance with the other operating parameters such as pH, temperature and flow rate. In the EC process, current density and the types of the anions define the current efficiency[17].

2.5 Effect of pH

The pH of the solution determines the conductivity of the solution, the electrode dissolution, and formation of hydroxides in the electrocoagulation process. Various empirical studies show that at high current efficiency using aluminum electrodes occurs at either acidic or alkaline condition than at neutral conditions. The nature of the pollutants determines the EC efficiency, however near pH 7 pollutant removal was found best. And, at neutral pH, the power consumption was high due to conductivity variations. pH effect is not significant at high conductivity. EC efficiency using aluminum electrodes decreases at pH 10 [18]. The optimal pH for wastewater treatment by electrochemical coagulation ranges from 6.5 to 7.5.

The pH of solution varies during electrocoagulation method and the final pH of the effluent significantly influences the pollutant removal efficiency.

2.6 Effect of Temperature

Temperature significantly influences the pollutant removal efficiency by using EC process. Temperature can have a positive or negative effect on electrochemical coagulation process, therefore in an EC process which is carried out at ambient temperature, it is necessary to know the factors that cause variations in temperature during the process. The increase in temperature decreases the pollutant removal efficiency due to the decrease in metal hydroxide formation [18]. At low temperature pollutant removal efficiency is also low due to the low anodic dissolution rate[19].

Temperature influences the EC process by altering the rate of reactions, solubility of metal hydroxides, liquid conductivity, and kinetics of gas bubbles, or small colloidal particles. Effect of temperature (20-50°C) on phosphate removal from wastewater using aluminum electrodes showed that increase in temperature increases the removal efficiency due to the increased mass transfer of aluminum ions from anode surface to bulk solution and increase in the rate of aluminum hydroxides formation. Increase in temperature above 50°C is not preferred as it is uneconomic and alters the rate of reaction and removal efficiency.

2.7 Effect of Initial Pollutant Concentration

The initial pollutant concentration is also considered as one of the effective parameters in pollutant removal by electrochemical coagulation. According to literature an increase in initial pollutant concentration (by keeping other parameters constant) reduces the pollutant removal efficiency of EC process. This is due to the circumstance that at fixed operating parameter the amount of coagulant generated will be fixed which is insufficient to form floc with high pollutant concentration. Therefore a longer electrolysis time is required to reduce the residual pollutant concentration up to the desired removal yield. Pollutant concentration not only decreases the EC efficiency it also increases the energy consumption by increasing the resistance and conductivity of solution.

3. Kinetics of EC Process

The kinetics of pollutant removal by electrochemical coagulation can be exhibited by the estimation of coagulant dose using an adsorption phenomenon. Kinetic studies are necessary to determine the dependency of pollutant removal rate on the operation parameters. The amount of pollutant (Qt) removed at time t is calculated by using the Equation $Qt = (C_0 - C_t)\ v\ w$

Where C_0 (mg/L) is the initial pollutant concentration and C_t (mg/L) is the pollutant concentration at time *t* in the aqueous phase, v is the solution volume (L), and w is the mass of metal hydroxide calculated. The mechanism of the adsorption of the pollutant on in situ-generated metal hydroxide process can be described by applying different kinetic models such as pseudo-first order, pseudo-second-order, Avrami and Elovich models.

3.1 Operating Cost Analysis

In the Electrocoagulation process, it is very necessary to achieve highest removal efficiency with minimum operating cost. The operating cost of electrochemical coagulation process depends on the cost of the electrode, electrical energy cost, sludge disposal, and fixed cost. The operating cost can be calculated by the following equation

$$\text{Operating cost} = a\,C_{\text{Energy}} + b\,C_{\text{Electrode}}$$

Where C_{Energy} and $C_{\text{electrode}}$ is the amount of electrical energy and electrode consumed per liter of wastewater treated, which is experimentally achieved. The unit price of electrical energy “a” and electrode material “b” can be obtained by the market.

The amount of electrical energy consumed per liter wastewater treated can be calculated by the given equation.

$$E_{EC} = UIt_{EC}$$

Where $E_{EC}$is the electrical energy in kWh/L, U the cell voltage in volt (V), I the current in ampere (A) and $t_{EC}$is electrolysis time.

The amount of electrode consumed (E_{EL}) per liter wastewater treated can be calculated by the given equation

$$E_{EL} = \text{Initial Weight of Electrode} - \text{Final weight of Electrode}$$

Table 1. Recent Application of EC in the Treatment of Wastewater

Sl. No	**Author and year**	**Waste water type**	**Anode/ cathode**	**Reactor type**	**Electrode arrangement:**	**No of electrodes:**	**Sample volume in (ml)**	**Optimum electrode gap in (cm)**	**Optimum current density**	**Optimum detention time in (min)**	**Temperature**	**pH**	**Initial pollutant Level (mg/L)**	**Optimum removal efficiency in (%)**
1	Tyagi et al. 2014[20]	Textile wastewater	Fe	Batch	-	2	1500	4	14-17 mA/cm^2	20	-	8.5	COD=600-650	COD=76 Colour=95
2	Farshi et al 2013.[7]	Distillery wastewater	Al SS	Batch	-		200	1	2	180	-	4	-	COD=70-72 Colour= 97-98
3	Akbal et al 2011.[21]	Metal plating wastewater	Al/Al Al/Fe Fe/Fe Fe/Al	Batch	Monopolar	6	650	1	10 mA/cm^2	20	-	9	Ni=394 Cu=45 Cr=44.5	Ni=100 Cu=100 Cr=100
4	Katal et al 2011.[22]	Paper mill wastewater	Al Fe	Batch	-	-	1500	1	70 mA/cm^2	30	20^{0}C-60^0 C	5-7	COD=1700 BOD=850 TOC=910 TSS=1060 TS=9801 Phenol=34 Colour=NM	COD=86(Fe) Colour=92(Al) Phenol=96 (Al) BOD,TOC,TS,TSS=NM
5	Kobya et al. 2011.[23]	Potable water	Al Fe	Batch	-	-	650	1.3	2.5 A/m^2	4(Al) 2.5(Fe)	-	7(Al) 6.5(Fe)	As=150mg/l	As =93.5 (Al) As = 94.1(Fe)
6	Kobya et al. 2011.[24]	Drinking water	Al Fe	Batch	MP-P	4	560	1.3	2.5 A/m^2	12.5(Fe) 15 (Al)	-	6.5(Fe) 7 (Al)	As =75-500mg/l	As=93.5(Fe) As= 95.7(Al)
7	Linares Hernandi et al 2009.[12]	Industrial wastewater	Al,Fe (Al+Fe)	Batch	MP	2	4000	2	45.45 A/m^2	60	-	8	COD =1700-2500, BOD$_5$=900-930 Colour(pt-C$_0$)=2500-3700 Turbidity(NTU) =1400-1800	COD=>99 BOD$_5$=>99 Colour=100 Turbidity=100
8	Kobya.et al 2010.[25]	Electroplating rinse wastewater	Fe	Batch	MP-P	2	650	1	30-60 A/m^2	30-80	25^{0}C	8-10	Cd=102 Ni=1175 Cy=120-261 COD=180-220	Cd=99.4 Ni=99.1 Cy=100 COD=NM

9	Zodi et al 2010[3].	Industrial textile wastewater	Al	Batch	-	2	2200	2	60-140 A/m^2	30-90	20^0C	7	COD=1260 Turbidity=1310[NTU] TS=1750	COD=70 Turbidity=90 TS=50
10	Kushwaha et al 2010.[5]	Dairy wastewater	Fe	Batch (synthetic)	BP	-	1.5	1	270	50	-	6-8	COD=3900 Turbidity=1744 NTU TS=3090 TN=113 Chloride=31	COD=70 Turbidity=100 TS=48.2 TN=92.75
11	B M Krishna et al 2010.[6]	Distillery wastewater	Al	Batch	-	2	1500	2	0.03-0.01A/cm^2	120	-	3	COD=42240-46440 BOD_5=6757-8600 BOD_5/COD=0.15-0.19	COD=72.3 BOD_5/COD=0.15-0.68
12	Vasudevan et al 2010.[19]	Drinking water containing boron	Mg SS	Batch	-	2	900	0.5	0.2Ad/m^2	30	±2K	7	Boron=3-7	Boron =86.32
13	Maghanga et al 2009.[26]	Tea factory wastewater	Steel	Batch	-	2	400	0.5	24V	NM	Above 20^0C	6	COD=293-607 BOD_5=42-193 Colour=2004-9210[Pt/Co]	COD=96.6 BOD_5=84 Colour=100 Electric conductivity= 31.5
14	Kalyani,balasubramanian et al 2009.[27]	Pulp and paper industrial effluent	Steel Al	Batch	-	-	-	1.5	NM	NM	-	NM	COD=32000 BOD=8225 BOD/COD=0.26	Colour=92-84 COD=95-89
15	InoussaZongo et al 2009. [4]	textile wastwater	MS Al Fe	Batch	-	2	$1cm^3$	NM	50-200A/m^2	60	20^0C	7.5	COD=1787 Turbidity=115 Conductivity=28	COD=74-88
16	Kobya et al 2008.[28]	Baker's Yeast wastewater	Al Fe	Batch	MP-P	4	800	2	70 A/m^2	50	20^0C	7	COD=2485 TOC=1061 Turbidity=2075 NTU TSS=503	COD=71(Al)-69(Fe) TOC=53(Al)-52 (Fe) Turbidity=90(Al) -56(Fe)

17	Tir&Moulai-Mostefa et al 2008. [29]	Industrial oil-in-water emulsion	Al/SS	Batch	MP-P	3	400	1	25 mA/cm^2	22	20^{0}C-22^{0}C	7	COD=62300 Turbidity=29700 [NTU]	COD=90 Turbidity=99
18	Kongjao. et al.2008[30]	Tannary wastewater containing organic and inorganic pollutants	Fe	Batch	MP-P MP-S BP-P	6	3000	5	15.7-24.6 A/m^2	20	30^{0}C	7-9	COD=4100-6700 BOD=630-975 Cr=11.5-14.3 TSS=600-955 Oil &Grease638-780	COD=95 BOD=96 Cr=100 TSS=96 TDS=50 TKN=62 Oil &Grease=99
19	Ilhan,kurt et al 2008.[31]	Leachate wastewater	Al Fe	Batch	Parallel	2	0.5	6.5	348-631 A/m^2	30	-	NM	COD=12860 BOD$_5$=5270 BOD$_5$/COD=0.41 Ammonia=2240 Turbidity=1340 Chloride=3100	COD,NH$_4$-H=56 (Al) COD,NH4-H=35 (Fe)
20	Sevilveli et al 2008.[32]	municipale solid waste leachate	Fe-Al	Batch (laborata ry scale)	-	-	2000	0.3	2-15mA/cm^2 (Al) 1-10 mA/cm^2 (Fe)	15	21^{0}C-22^{0}C	9	COD=4022 TOC=1295 Conductivity=25.11 Colour(Hz)=2650	COD=56-73(Al^{3+}), 87-90(Fe^{2+}) TOC=46-49(Al^{3+}), 58-68(Fe^{2+}) Colour=69-76(Al^{3+}),86-99(Fe^{2+})
21	Roa-Morales et al 2007.[33]	Pasta and cookie proccessing wastewater	Al	Batch(pilot scale)	-	8	1.5	-	18.2 mA/m^2	60	-	4	COD=7500 BOD$_5$=3445 Colour=35Pt-Co Turbidity=1153	COD=90 BOD$_5$=96 Colour=57 Turbidity=97 TS=95
22	Alaadin A et al 2008. [34]	Muncipal wastewater	stainless steel	Batch	-	2	1200	3	0.8A	5	-	7	TSS=126-160 Turbidity=49-53 Total BOD=84-112 Soluble BOD=26-32 Particulate BOD=51-84 Chloride=1238 Conductivity=4000	TSS=95.4 BOD=99

23	Kobya et.al 2006.[35]	Potato chips manufacturing wastewater	Al Fe	Batch	MP	4	250	1.1	20-300 A/m^2	5-40	22^{0}C	4-6	COD=2200-2800 Turbidity:260-610 NTU BOD=1650-2150	COD=60 Turbidity=98 BOD=NM
24	NN Rao et al 2001.[36]	tannery wastewater	Ti/mno2 -titanium	Batch	-	2	60cm³	4	0.1-0.6A	20	-	8	Colour=1100-1150 Pt-C$_o$ COD=480-550 Chloride=4500-4600 Ammonia=100-110	Colou=75-95 COD=50-75 Ammonia=96

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