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Electro- Fenton Process for Waste Water Treatment A Review

Pushpalatha M[1]
[1]*M.Tech Scholar, Department of Environmental Engineering, Sri Jayachamarajendra College of Engineering, Mysuru-570006, Karnataka, India.*
pushpausha474@gmail.com

Krishna B M[2]
[2]*Associate Professor, Department of Environmental Engineering, Sri Jayachamarajendra College of Engineering, Mysuru-570006, Karnataka, India*
bmkrishna_71@yahoo.com

Abstract – In recent year large amount of water and wastewater generated from various industries and produce organic matters and contaminated the water, this organic this organic compound in water poses serious problems to public health as well as environment. The advanced oxidation process (AOPs) is one of the advanced treatment method for wastewater remediation. In AOPs, Electro- Fenton process (EFP) is one of the best methods for the wastewater treatment. In this present research article review the treatment of different wastewater by using EFP and this review paper also focus on the development of EFP, application and advantages of EFP for the remediation of wastewater from various industries and some affecting factors like pH, temperature, Electrode distance, current density, Fe^{2+} Concentration, H_2O_2 Concentration etc., and this review concluded that compare to other methods EFP is a promising method for organic treatment and its is also environmental friendly. Therefore electro Fenton process is one of the best methods in AOPs.

Keywords – Advanced oxidation process, Electro-Fenton technique, wastewater, current density, Fe^{2+} Concentration, H_2O_2 Concentration.

I. INTRODUCTION

In recent year large amount of water and wastewater generated from various industries like tannery, pulp and paper industry, dairy industry, steel industry, tyre and tube manufacturing industries etc., Nowadays more organic contaminants from agricultural runoff, soil contaminants, landfill leachates, industrial wastewater etc., this organic compounds in water poses serious problems to public health as well as environment. All over the world the present challenging problem facing is water quality and availability of water (1). Various treatment methods are discovered to focus on waste minimization and water conservation in the recent years, in that Advanced Oxidation Processes (AOPs) is one among them. AOPs refer to the chemical process which follow oxidation and helps to degrade biologically toxic and non degradable chemicals.

II. FENTON PROCESS

In AOPs, oxidation using Fenton's reagent is one of the best effective, alternative, powerful and environmental friendly technique, this method is used for the degradation of a large number of hazardous and organic pollutants. fenton reagent first named as fenton chemistry, fenton chemistry is the oxidation of organic substrate by Iron (II) and hydrogen peroxide. It was first described by Henry John Horstman Fenton in 1890. The first fenton process reported by Fenton was Maleic acid oxidation. Oxidation, Neutralization, Flocculation and sedimentation are the main steps involved in the Fenton process. The degradation mechanism of organic pollutants by Fenton reaction is given in eqs 1 to 4

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + HO^\cdot \quad (1)$$

$$RH + HO^\cdot \rightarrow R^\cdot + H_2O \quad (2)$$

Where, RH denoting organic pollutants

$$R^{\cdot} + Fe^{2+} \rightarrow R^{+} + Fe^{2+} \quad (3)$$

$$Fe^{2+} + HO^{\cdot} \rightarrow Fe^{3+} + OH^{-} \quad (4)$$

The Fenton process has some of the disadvantages in the treatment process because it require high concentration Fe^{2+} and large amount of Fe sludge is formed so that wastewater treatment is expensive and it needs large amount chemicals and manpower. Fenton's reaction is only limited for narrow pH because iron ions will be precipitated at higher pH values and storing and transport of H_2O_2 are difficult. To overcome this above problem, Electrochemical advanced oxidation processes (EAOPs) based on Fenton's reaction (1).

III. ELECTRO FENTON PROCESS

EAOPs are eco – friendly methods and recent days it received more attention for water remediation. Electro – Fenton is the most popular process in EAOPs (2). , Electro – Fenton process is a promising technology for wastewater treatment process and its more economical, efficient, and environmental friendly for treatment and removal of organic matter compared to conventional technologies (3). This technology based on the continuous electro generation of H2O2 at a suitable cathode by the reduction of dissolved oxygen or air along with the addition of an iron catalyst to the treated solution to produce oxidant $^{\cdot}OH$ at the bulk via Fenton's reaction.

$$O_2 + 2H^{+} + 2e^{-} \rightarrow H_2O_2 \quad (5)$$

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^{-} + HO^{\cdot} \quad (6)$$

$$Fe^{3+} + e^{-} \rightarrow Fe^{2+} \quad (7)$$

Electro – Fenton process solves some problems generated from conventional Fenton process, for example degradation of organic pollutants increase because of the continuous regeneration of Fe^{2+} at the cathode and H_2O_2 is produce at onsite so that it avoids storage and transport problems (1).

The typical mechanism of the electro Fenton process illustrated in the below figure 1. Electro Fenton Process includes both electrochemical and Fenton treatment methods and each method are powerful and effective method for the treatment process.

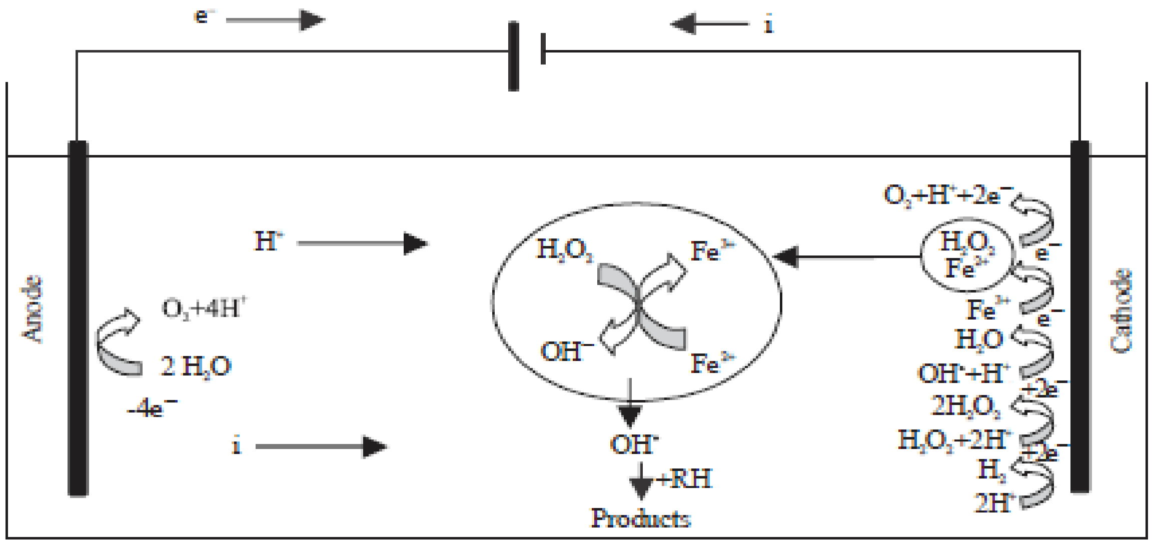

Fig 1 Illustration of the reaction mechanism of electro – Fenton process
(source: Qiang, 2002)

Compare to conventional Fenton's Reagent or other treatment technology the more attention has been paid to Electro Fenton Process (EFP) for various wastewater treatment (4). Compare to conventional Fenton's reagent EFP is effective, environmental friendly, easy to treat, react well with organics and it doesnot produce secondary pollutants and toxic compounds during oxidation but it is also have some disadvantages like EFP should be maintained in acidic conditions, H_2O_2 presence may harmful to micro – organisms, during Fenton process large amount of small flocs were observed that flocs take more or large time to settle, H_2O_2 concentration higher than 1000 mg/l that effects on the biodegradability of the wastewater because of some residual H_2O_2

IV. APPLICATIONS

In recent days the application of Electro – Fenton process is focused more on the remediation of water and wastewater contaminated by several organic pollutants such as dyes, pesticides and herbicides, phenolic compounds, leachates, drugs etc (2)(3) the applicability of Electro – Fenton process for decolourization, removal of odour ingredients with good energy efficiency, EFP is applied for destruction of toxic waste and non – biodegradable effluents and it is more suitable for secondary biological treatment. Some application of electro – Fenton's process and research highlights as shown in the below table 1.

Table 1: Application of electro – Fenton's process and research highlights

Sl No	Application Field	Research highlights	Reference
1	Degradation of Phenol – containing wastewater	• Improved EF – Fere method used • initial ferric ions concentration was 800 mg/l and current density was 0.75 A • higher COD removal efficiency obtained at initial ferric ion concentration, current density at 1 A and 1.2 Q_T H_2O_2 • Stainless steel fiber can be used as a cathode and it is good cathode electrode for EFF process	5
2	Treatment of Tissue paper Wastewater	• Electro – Fenton Method is adopted for Tissue paper wastewater • Initial COD concentration of 1200 mg/l was treated • removal efficiency is 80% after 60 min of running and current density of 20 mA/cm^2	6
3	Removal of Chromium from Aqueous solution	• Electro – Fenton process is used for chromium in aqueous solution • optimum pH 5, voltage peroxide dosage is 30V and 50 mL/L	4

V. AFFECTING FACTORS

pH

pH played a very important role in EF process and it is a significant impact in EF process because it control the variation of iron and hydroxyl radical generated. In lower pH electro Fenton process occurs more so that all experiments carried in the acidic range. The reaction gets slow down because of complex species of Iron and it is slowly reacts with peroxide compared to $[Fe(OH)(H_2O_5)]^{2+}$. In lower pH, the EF process may occur more efficient and the pH solution decreases the conductivity increases (6). In pH range 3 – 4 the maximum amount of waste removal has been observed. The pH solution below 2, the waste degradation has been reduced because of the generation of $[Fe(H_2O)_6]^{2+}$ and $[H_3O_2]^{2+}$ and the pH solution above 4 the insignificant amount of waste degradation occur due the formation of hydroxyl radical (7). The maximum removal efficiency occurs in pH range 3 and the pH value increases with the increase in the reaction time (4). At pH range 3 the reaction time is inversely proportional to the electrical power used (8). A low pH is more favorable for the production of hydrogen per-oxide so that the conversion of DO to hydrogen peroxide consumes protons in acidic solution (9). At pH value is high the iron precipitates and it reducing the concentration of the dissolved iron (11). The pH value increases, the iron ions precipitates. If pH is less than 2, H_2O_2 cannot be discomposed to $OH^{.}$ by Fe^{2+} (5). According to all research it has been confirmed that the optimum pH of Fenton process is around 3 (2). The redox system and decolourization gives more stable results when pH is less than 3.5, Ferric ions are easily formed because of ferrous ions and it is propensity to produce ferric hydroxo complex and H_2O_2 is unstable and it is easily decomposes itself in basic solutions.

The different pollutants removal by EF process the different optimum pH is necessary and it is reported in Table 2. From Table 2, it was found that the optimum pH for EF process varies from 2 to 4.

Table 2: Optimum pH values of electro Fenton process in various researchers.

Sl No	Pollutant	Electrodes	Optimum Ph	Efficiency (%)	Time (min)	Reference
1	Tissue paper wastewater	Cathode: cylindrical iron electrochemical reactor Anode: turbine impleller with 8 flat blades	2	COD – 80	-	6
2	Dye wastewater(Alizarin Red)	Cathode: Graphite felt piece – 60cm^2 thickness – 0.5cm Anode – Cylindrical grid	3	TOC – 95%	210	11
3	Landfill Leachate	Anode: Ti/Iro_2-RUO_2 – TiO_2 – 10cm×15cm Cathode:Titanimum - 10cm×15cm	3	COD - 67.7%	240	12
4	Synthetic wastewater	Iron elecrodes	3	97	25 min	4
5	Real dyeing wastewater	Cathode: graphite rod Anode:Pt/Ti	3	70.6	150 min	9

Temperature

Temperature plays a significant role in the electro Fenton process. The temperature is negatively affected the removal efficiency. The removal efficiency decreases when temperature increases. The concentration of H_2O_2 decreases, the temperature increases because of the reduction in the DO concentration. Rate of self decomposition of H_2O_2 to water and oxygen increases with temperature. The decomposition rate doubles when temperature rises to 10° C. The negative impact on the process efficiency when temperature is too low and too high. The optimum range of temperature between 20 and 30° C because of relatively higher treatment efficiency (9)(12). The optimum temperature and corresponding removal efficiencies reported by various studies are given in table 3

Table 3: Optimum Temperature of EF process in various studies

Sl no	Pollutant	electrodes	Experimental condition	Temperature (° C)	Removal efficiency (%)	Reference
1	Dye wastewater(Alizarin Red)	Cathode: Graphite felt piece – 60cm^2 thickness – 0.5cm Anode – Cylindrical grid	pH – 3 Current density – 300 mA Fe^{2+} - 0.2 mM Time – 210 min Rotation rate – 700 rpm	25	TOC – 95%	11
2	Real dyeing wastewater	Cathode: graphite rod Anode:Pt/Ti	pH – 3 Current density – 68 A/m^2 Time -150 min Fe^{2+} - 15 mM Oxygen sparing rate – 0.3 dm^3/min	25	70.6	9

Applied Current Density

Current density plays a very important role in EF process. Energy consumption increases with increasing the current density. The current density increases the removal efficiency also increases (6). The production rate of hydrogen peroxide on the cathode increases when the current density increases. The removal efficiency increases with the current density (9). The applied current is directly related to the cost of EF process. The degradation of substrate increases in the beginning because of the increase in current density. In electro Fenton process the current density is always low. In EP process large reaction rate occurs because of the higher current density. According to some research works the current density in the EF process should less than 10 A/m^2 (5).

The optimum applied current and the corresponding removal efficiency of EF process from various studies as shown in table 4

Table 4: Optimum current density by EF process from various research works

Sl No	Pollutants	Electrodes	Experimental conditions	Applied current density	Removal efficiency	reference
1	Dye wastewater(Alizarin	Cathode: Graphite felt	pH – 3	Current density –	TOC – 95%	11

	Red)	piece – 60cm^2 thickness – 0.5cm Anode – Cylindrical grid	Fe^{2+} - 0.2 mM Time – 210 min Rotation rate – 700 rpm Temp - 25° C	300 mA		
2	Real dyeing wastewater	Cathode: graphite rod Anode:Pt/Ti	pH – 3 Time -150 min Fe^{2+} - 15 mM Oxygen sparing rate – 0.3 dm^3/min	Current density – 68 A/m^2	70.6	9

Fe^{2+} Concentration

Fe^{2+} Concentration is an important affecting factor in the EF process. If Fe^{2+} Concentration increasing, the hydroxyl radical generated via Fenton's reaction. Fe^{2+} Concentration increases, the removal rate declined. Fe^{2+} Concentration is higher than 0.2mM, the percentage of hydroxyl radicals from Fe^{2+} ions increased (1). The EF processes can operate in optimum conditions with small initial concentration of catalytic Fe^{2+}. TOC removal efficiency increasing, the Fe^{2+} Concentration also increases (13). The degradation rate decreases by increasing the Fe^{2+} Concentration. The Fe^{2+} Concentration effects on the kinetic rate also. Initial Fe^{2+} Concentration is beneficial for the Fe^{3+} - H_2O_2 complexes (11) (14). If Fe^{2+} Concentration is more than 500 mg/l, OH radicals are scavenged through the reactions

$$Fe^{2+} + OH \rightarrow Fe^{3+} + OH^-$$

The Fe^{2+} Concentration is higher, the OH radicals amount availability to oxidize organic matter become low and Fe^{2+} Concentration increases the COD values decreases. The optimum Fe^{2+} Concentration and Removal efficiencies from EF process as shown in table 5

Table 5: The optimum Fe^{2+} Concentration and Removal efficiencies from EF process from various studies

Sl No	Pollutants	Electrodes	Experimental conditions	Fe^{2+} concentration	Removal efficiency	reference
1	Dye wastewater(Alizarin Red)	Cathode: Graphite felt piece – 60cm^2 thickness – 0.5cm Anode – Cylindrical grid	pH – 3 Current density – 300 mA Time – 210 min Rotation rate – 700 rpm	Fe^{2+} - 0.2 mM	TOC – 95%	11
2	Real dyeing wastewater	Cathode: graphite rod	pH – 3 Current density	Fe^{2+} - 15 mM	70.6	9

		Anode:Pt/Ti	– 68 A/m^2 Time -150 min Oxygen sparing rate –0.3 dm^3/min			

H_2O_2 Concentration

H_2O_2 Concentration is one of the most important factors in EF process and plays a major role for COD removal efficiency in EF processes. H_2O_2 Concentration dosage increases, the hydroxyl radicals are also increased. H_2O_2 Concentration dosage increases, the COD removal rate also increases (6). The removal efficiency of TOC increases with increasing the H_2O_2 Concentration. The lower the concentration of H_2O_2 does not influence the rate of removal of TOC (13). The removal efficiency of pollutants increases due to increase in the hydroxyl radical's concentration as a result of addition of H_2O_2. H_2O_2 Concentration dosage is high in the removal efficiency because of hydroxyl radicals scavenging effect of H_2O_2 Concentration and the recombination of the hydroxyl radical (14). EF process increases with increasing the H_2O_2 Concentration, the removal efficiency of chromium is also further improve the quality and H_2O_2 Concentration presence helps to faster production of Fe^{3+} (4).

Distance between the electrodes

The distances decreases between the electrodes cause to decrease of the ohmic drop through the electrolyte and then an equivalent decreases the electrical conductivity and voltage. Closes the electrodes gives the better performances. To avoid the short circuits between anode and cathode, the electrodes should keep approximate distance (5). The electrode distance between 1.3 and 2.1 cm, the COD removal efficiency from landfill leachate remains same and the removal efficiency of EF process system is less when the electrode distance is shorter and larger (2).

The following table gives about the electro Fenton process and its research highlights. Some of the review papers discussed here

Table 6: Electro- Fenton process and its highlights

Sl No	Wastewater type or pollutant	Research Objective	Reactor	Electrodes	Electrode distance	Initial pollutant conc	Optimum condition	Volume of treated wastewate	Removal efficiency	Reference
1	Dye wastewater(Alizarin Red)	The removal of the anthaquinone dye alizarin red was investigate by using EFP in this research work	Undivided cylindrical cell (dia-7.4cm and 0.25 dm^3 volume)	Cathode: Graphite felt piece – 60cm^2 thickness – 0.5cm Anode – Cylindrical grid	1.6 cm^2	-	pH – 3 Current density – 300 mA Temp -25° C Fe^{2+} - 0.2 mM Time – 210 min Rotation rate – 700 rpm	200 $mgdm^{-3}$	TOC – 95%	11

2	Landfill Leachate	The effects of Initial pH, current density, PDS and ferrous ion dosage on COD removal was investigate in this research	Electrolytic reactor (plexy glass) with 1000 mL capacity	Anode: Ti/Iro_2-RUO_2 – TiO_2 – 10cm×15cm Cathode: Titanimum - 10cm×15cm	2 cm	1900 mg/l	pH – 3 PDS – 62.5 mM Fe^{2+} - 15.6 mM Current density – 13.89 mA/cm^2 Reaction time –240	1000 mL	COD - 67.7%	12
3	Tissue paper wastewater	The effects of initial pH, current density and concentration of H_2O_2 on removal efficiency and energy consumption was investigate in this study	Electrochemical reactor	Cathode: cylindrical iron electrochemical reactor Anode: turbine impleller with 8 flat blades		1200 mg/l – COD	pH – 2 current density – 20 mA/cm^2 concentration of H_2O_2 - 0.2M	0.5 L	COD –80%	6
4	Dimethylaniline solution	The effects of initial pH, current density and concentration of H_2O_2 was determined in this research work	Cylindrical reactor (radius – 6.5 cm, ht – 35 cm) with 3.5 L capacity	Anode: Titanium metal coated with RuO_2/ IrO_2 Cathode: Stainless steel	-	-	pH- 2 Fe^{2+} - 1mM H_2O_2 – 20mM Current density – 7.6 A/m^2	3.5L	TOC – 60%	14
5	Synthetic wastewater	The effects of EFP using iron electrode on the removal of chromium from aqueous solution has been investigated in this study	Batch plexiglass reactor with 1 L capacity	Iron elecrodes	2cm	100 – 1000 mg/L	pH – 3 voltage – 30V H_2O_2- 50 mL/L Time – 25 min	1L	97%	4

6	Real dyeing wastewater	Removal of color from real dyeing wastewater by EF technology using a three – dimensional graphite cathode is the main objective of this research work	Small glass beads reactor with 0.3 cm in diameter	Cathode: graphite rod Anode:Pt/Ti			pH – 3 Current density – 68 A/m^2 Time -150 min Fe^{2+} - 15 mM Temp -25°C Oxygen sparing rate – 0.3 dm^3/min	1.5dm^3	70.6%	9
7	Real wastewater	The main objective of this study was to treat real wastewater from sustainable environmental research center	Cylindrical pyrex tube	Anode: titanium rod coated with Iro_2/RuO_2 Cathode: Cylindrical Ti DSA		COD- 24000 mg/L TOC – 16500 mg/L	pH – 2 current density – 20A Fe^{2+} - 2000mg/l H_2O_2 (50wt %) – 6 mL/min Time – 2520 min	4L	TOC – 98% COD – 97%	15
8	Leather tannery industrial wastewater	The reduction of COD in wastewater from leather tannery industry by using electro Fenton process was investigate in this research work	Glass electrode	Iron electrode	6cm	-	pH – 3, 5, And 7.2 time 10 min	0.41 L	60 – 70% of COD	8
9	Phenol containing wastewater	The main objective of this research work is the degradation of phenol containing	Cylindrical electrolytic cell	Cathode: stainless steel Anode: RuO_2, SnO_2 and	-	800 mg/l	pH – 3 current density – 1.0 A H_2O_2 – 1.2 Q_T	-	78%	5

		wastewater using an improved EF process		pbO_2						
10	Synthetic dye wastewater	The main objective of this research work to analyse the recent advances in the application of EFP for removal of synthetic dye in wastewater	Glass cylindrical reactor with 0.675 L	Graphite and stainless steel	240.4mm	-	pH – 2 Fe^{2+} - 1.0 mM	0.675 L	Lissamine greem and Methyl orange – 80% Reactive black 5 – 60%	16
11	Azo dye wastewater	The main objective of this research work is to know about the degradation of azo dyes in water by using EF process	Cylindrical cell	Anode: Pt Cathode: Carbon felt	-	-	pH - 2	110 mL	COD - 80%	17
12	Aniline solution	The main objective of this research work is to identify the effects of the feeding mode and concentration of hydrogen peroxide on the aniline solution by EF process and Fluidized – bed fenton process	Acrylic reactor (15cm×21cm×20 cm) with 5L capacity	Anode: RuO_2/IrO_2 – coated with titanium rod Cathode: stainless steel	-		0.01M aniline solution Fe^{2+}- 1.07×10^{-3} SiO_2 – 74g/L Current : 4A pH – 3.2 time – 60 min	-	Aniline – 96% TOC – 20 – 30%	18

13	Propham aqueous solution	The main purpose of this research work was identify the efficiency of EF process by using different electrodes and also investigate the effects of electrode materials on the accumulation of the oxidation	Undivided cylindrical glass cell – 0.175 L	BoronDopedDiamond, platinum, Carbon Sponge	-	-	Propham – 0.5 mM Current density : 100mA pH : 3 temp : 35° C Fe^{3+}- 0.2 mM Time : 30 min	0.15L	81%	19
14	Acetonitrile aqueous mixture	The main objective of this research work was to remove the 17β – Estradiol in acetonitrile aqueous solution by using EF process	-	Cathode: carbon felt piece(80 cm^2) Anode: platinum electrode (16 mm^2)	-	-	pH – 3 current density : 200 mA Fe^{2+} conc : 0.2 mM	250 mL	100%	20
15	Azo dye wastewater	The research work aim was to investigate the experimental design methodology to identify the influence of the experimental parameters on degradation of direct orange 61 by using EF process	Undivided cylindrical glass cell	Anode: Pt Cathode: carbon felt	-	-	pH – 3 current density – 60 mA Fe^{2+} concentration – 0.1 mM Time: 6h	500 mL	TOC – 98%	21

16	Rhodamine B	The research aim was to design 3-D EF system to remove RhB in wastewater	Rectangular 3-D electrode reactor (10cm×15cm×4cm) with 200 mL capacity	Cathode: carbon fiber Anode: Ti/RuO_2 – IrO_2 Foam nickel was used as particle electrodes	-	-	pH – 6.2 applied voltage – 2 V Fe^{2+}concentration – 3 mmol/l	200 mL	RhB – 99%	22

CONCLUSIONS

During rapid industrialization, the large amount of wastewater generated and contaminated. The recent advances have investigated the different methods for wastewater treatment. In that advanced oxidation process is advanced method for the wastewater treatment, among the different AOPs several researchers have demonstrated that EFp is a promising method and it is more economical, efficient, environmental friendly to remove organic matters. This review paper also concluded that EFP is good for removal of COD, TOC, Color and it is alternative method for treatment of wastewater containing synthetic dyes due to this efficient and low operating cost and environmental friendly method

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