Matured Landfill Leachate treatment by Electro Fenton and Photo Electro Fenton process

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Abstract— *Landfill leachate treatment is an integral part of municipal solid waste management. A novel approach for management of landfill leachate by Electro fenton process is researched here for the feasibility of biological treatment. Landfill leachate if irradiated by means of ultraviolet light along with Electro fenton process (EF), can accelerate the degradation of organic compounds. Landfill leachate was subjected to UV irradiation using UV lamp and sun light along with Electro fenton process. Use of sunlight is an inexpensive and renewable energy source in the solar photo electro-Fenton (SPEF) process. Percentage removal efficiencies of landfill leachate pollutants -TSS, BOD, COD, Ammonia Nitrogen, Phosphate, Sulphate, Sulphide and Chloride obtained after PEF process with UV light is 85.4, 58.6,75.4, 50.1, 100 , 58, 89 &60 , and that obtained after SPEF process is 79.6, 53, 70.4, 47.5, 90.5, 51 ,83.6 & 52. From the results PEF with UV lamp is more efficient than Solar photo electro-Fenton (SPEF) process. Though the degradation of pollutants by PF and SPEF processes are greater than EF process, the biodegradability index of landfill leachate enhanced to 0.36 and 0.38 respectively. The biodegradability index of landfill leachate treated by EF process enhanced from 0.21 to 0.43. Hence Electro Fenton process can be used as a pretreatment method before biological process for matured landfill leachate.*

Key words: Landfill leachate, Electro fenton process, Photo Electro Fenton process, UV irradiation, Solar Photo Electro Fenton process

I. INTRODUCTION

Municipal solid waste keeps growing as a result of increasingly wealthy lifestyles and continuing industrial and commercial development in many countries around the world. Landfilling is the most frequently employed worldwide of all available dumping options under the solid waste management system.[1] Leachate generation is a major problem for municipal solid waste (MSW) landfills and management of landfill leachate has become one of main focus for the environment management of landfill. In general, leachates may contain large amounts of organic matter (biodegradable and refractory to biodegradation), where humic-type constituents consist an important group, as well as ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts. The characteristics of the landfill leachate can usually be represented by the basic parameters COD, BOD, the ratio BOD/COD, pH, Suspended Solids (SS) and Ammonium nitrogen (NH_3-N). [2][3]. The methods of leachate treatment can be divided into biological and physicochemical methods. Conventional biological treatment methods are inadequate to treat matured landfill leachate. They cannot completely remove all pollutants in the leachate. As age of leachate increases, it matures & non-biodegradable or recalcitrant substances become predominant. The leachate still contains many of pollutants after biodegradation, such persistent organic compounds. Advanced

Oxidation Processes (AOPs) are promising methods to treat effectively the recalcitrant substances present in landfill leachate.[4]

A. Advanced Oxidation Processes (AOPs)

Advanced Oxidation Processes (AOPs) have been successfully used as pretreatment methods in order to reduce the concentrations of toxic organic compounds that inhibit biological wastewater treatment processes.[5] In AOPs oxidation of organic contaminants occurs primarily through reactions with hydroxyl radicals. AOPs involve two stages of oxidation (1) the formation of strong oxidants (hydroxyl radicals) and (2) the reaction of these oxidants with organic contaminants in water. The main mechanisms of AOPs are the generation of highly reactive free radicals. Hydroxyl radicals (HO•) are effective in destroying organic chemicals because they are reactive electrophiles (electron preferring) that react rapidly and non selectively with nearly all electron-rich organic compounds. They have an oxidation potential of 2.33 V and exhibit faster rates of oxidation reactions comparing to conventional. Once generated, the hydroxyl radicals can attack organic chemicals by radical addition, hydrogen abstraction and electron transfer. These radicals are very reactive, attack most organic molecules, and are not highly selective. Thus AOPs are effective in decomposing many toxic and bio-resistant organic pollutants without producing additional hazardous by-products or sludge which requires further handling.

A great number of methods are classified under the broad definition of AOPs. Most of them use a combination of strong oxidizing agents (e.g H_2O_2, O_3) with catalysts (e.g. transition metal ions) and irradiation (e.g. ultraviolet, visible). Fenton‘s reactions seem to be some of the most popular technologies for wastewater treatment. [6]

Fenton’s reagent is a mixture of $FeSO_4.7H_2O$ (Catalyst) & hydrogen peroxide (oxidising agent). It will produce more OH• radicals and the oxidation of the organics to CO_2 can be enhanced. Hydroxyl radicals (•OH) have an oxidation capability of 2.8V and display quicker rates of oxidation responses when contrasted with conventional oxidants[7] This system is considered as the most promising treatment among AOPs for remediation of highly contaminated waters. Because of its simplicity, the Fenton reaction is the process most often applied when it is necessary to remove recalcitrant compounds. Fenton process can oxidize and mineralize almost all the organic carbons to CO_2 & H_2O.

$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH- + HO•$

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

$HO• + RH \rightarrow H_2O + R•$

$R• + Fe^{3+} \rightarrow R+ + Fe^{2+}$

It is found that the Fenton reaction can be efficiently enhanced in photo electro assisted Fenton process since Fe^{2+} may complex with certain target compounds or byproducts, produced by UVA light and current. The ferric complexes would be reduced to ferrous ion from the photo-reduction and by reduction in the cathode. This would induce the Fenton chain reaction efficiently [8] [9]. However, the major drawback of the Fenton reaction is the production of iron sludge waste.

B.Electro Fenton Process (EF)

To maintain a strategic distance from the impediments of conventional Fenton oxidation, for example, loss of reactivity of chemicals and sludge generation, an altered procedure called Electro Fenton process was developed [9]. This process is the use of electrically assisted Fenton reaction. It will produce more OH• radicals and the oxidation of the organics to CO_2 can be enhanced. In this experiment, H_2O_2 is added and Fe^{2+} is provided from sacrificial cast iron anodes. EF method has the advantage of allowing a better control of hydroxyl radical production. In electro-fenton process, soluble Fe^{3+} can be cathodically reduced to Fe^{2+}. The fast generation of Fe^{2+} accelerates the production of OH•.EF process has quick reaction rates

as contrasted and other chemical treatments. In addition, electricity as a perfect energy source and it is utilized as a part of the procedure, so the general procedure does not make any secondary pollutants [11].

The mechanism of EF process is represented in equations as below [12]

At anode:

$Fe\ (s) \rightarrow Fe^{2+}\ (aq) + 2e^-$

$Fe^{2+}\ (aq) + 2\ OH^-\ (aq) \rightarrow Fe(OH)^2$

At cathode:

$H_2O\ (l) + 2e^- \rightarrow H_2(g) + 2OH^-(aq)$

Overall:

$Fe\ (s) + 2\ H_2O\ (l) \rightarrow Fe(OH)_2 + H_2(g)$

Higher electro regeneration of ferrous ion from ferric ion with increasing current increases the efficiency of EF process. But the efficiency of EF process will be less at higher current density after a certain limit.

C. Photo Electro Fenton Process (PEF)

When EF process is irradiated by means of ultraviolet light, it can accelerate the degradation of organic compounds. The ferric complexes would be reduced to ferrous ion from the photo-reduction and by reduction in the cathode. This induces the Fenton chain reaction efficiently. Fe^{3+} ions generated are photo chemically transformed to Fe^{2+} ions. Fe^{2+} ions is improved when the system is illuminated with UVA light (λ = 320-400 nm).[13]

In PEF the ferric complexes would be reduced to ferrous ion from the photo-reduction and by reduction in the cathode. This would induce the Fenton chain reaction efficiently. When the EF process is irradiated by mean of UV light, this can accelerate the degradation of organic compounds by two principal pathways: a) the photolysis of Fe^{3+} -oxidation products complexes, and b) improving the Fe^{2+} regeneration from the photo reduction of Fe^{3+} ions according to the equation. [9].

$Fe(OH)^{2}+ + h\upsilon \rightarrow Fe^{2+} + HO\bullet$

D. Solar Photo Electro-Fenton Process (SPEF)

Sunlight can be used as the alternative for artificial UV irradiation. Use of this is as inexpensive and renewable energy source in the solar photo electro- Fenton (SPEF) process. The solar photo electro fenton process is considered like a project environmentally feasible because it does not cause or increase the ecosystem fragmentation and that does not exceed the basin load capacity where it intends to locate. When the EF process is irradiated by means of solar light, this can accelerate the degradation of organic compounds by two principal pathways: a) the photolysis of Fe^{3+} -oxidation products complexes, and b) improving the Fe^{2+} regeneration from the photo reduction of Fe^{3+} ions. [13][14]

II. MATERIALS AND METHODS

The landfill leachate samples were collected from Municipal Solid Waste Disposal Facility at Brahmapuram, Kochi. The various parameters (BOD, COD, chloride, sulphide, sulphate, phosphate, TSS, Ammonium nitrogen) were anlaysed as per standard procedure. The characteristics of original sample obtained are given in table 1. The BOD/COD ratio of leachate sample is 0.21.

Table 1 Characteristics of landfill leachate

Parameter	Unit	Average Concentration
pH	-	8.1
TSS	mg/l	498
COD	mg/l	23200
BOD	mg/l	4872
Sulphate	mg/l	587
Phosphate	mg/l	185
Sulphide	mg/l	20.1
Chloride	mg/l	2670
Ammonia nitrogen	mg/l	2196
BOD/COD ratio	-	0.21

A Electro-Fenton Process

The EF experiment was conducted in 1000ml beaker as reactor with cast iron plates as electrodes. The reactor used for the study was a 1000ml borosil glass beaker. 800 ml landfill leachate sample was used for batch studies. The reaction mixture was continuously stirred by a magnetic stirrer. Experiments were carried out at room temperature. Cast iron plates (12 cm x 6.5 cm x 0.1 cm) with maximum surface area that will be fit to the 1 L borosil glass beaker were used as electrodes. The electrodes were vertically positioned in the beaker. The distance between two electrodes was fixed 2.5 cm. The cast iron plates acted as the source of Fe^{2+} ions also. A pre-decided amount of hydrogen peroxide (H_2O_2) was added. The pH of solution was adjusted using H_2SO_4. A direct power supply was used to provide the desired current. The electric circuit consists of a regulated DC power supply, ammeter connected to the reactor. The Electrodes were connected to a DC power source. Fig 1 shows the experimental setup. 27 sets of experiments according to experimental design using RSM with varying operating conditions were performed. Samples were collected and analyzed for COD, Ammonia nitrogen, chloride, sulphate, sulphide and phosphate. The best operating conditions giving maximum pollutant removal has been found using RSM. Experiment was repeated with optimum operating conditions and the effluent was analyzed.

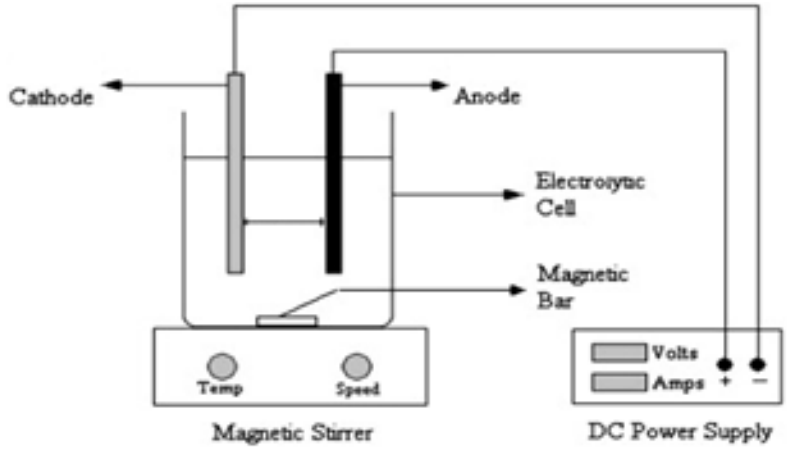

Fig 1. Schematic diagram of Electro-Fenton process

After 45 minutes treated samples were allowed to settle for 2 hours and the supernatant was used for analyzing the parameters.

B.Photo Electro- Fenton (PEF) Process

The reactor used for photo electro- fenton (PEF) process was a 1000ml borosil glass beaker. 800 ml leachate sample was used for batch studies. The reaction mixture was continuously stirred by a magnetic stirrer. Experiments were carried out at room temperature. The cast iron electrodes (12 cm x 6.5 cm x 0.1 cm) were placed vertically inside the reactor. The distance between two electrodes was fixed 2.5 cm. The cast iron plates acted as the source of Fe2+ ions also. The connections were made. The EF reactor was irradiated with UV light with 8W capacity. It was placed top of the glass beaker. It was covered with aluminium foil. The UV lamp was turned on. Reaction mixture is continuously stirred by a magnetic stirrer. The reaction mixture had H_2O_2 dosage 57.6 % , the current density 140.5 A/m^2 ,reaction time 45 min. and pH 2.9. After 45 minutes treated samples were allowed to settle for 2 hours and the supernatant was used for analyzing the parameters.

C. Solar Photo Electro- Fenton (SPEF) Process

The reactor used for solar photo electro- fenton (SPEF) process was a 1000ml borosil glass beaker. 800 ml synthetic leachate sample was used for batch studies. The reaction mixture was continuously stirred by a magnetic stirrer. Experiments were carried out at room temperature. The cast iron electrodes (12 cm x 6.5 cm x 0.1 cm) were placed vertically inside the reactor. The distance between two electrodes was fixed 2.5 cm. The cast iron plates acted as the source of Fe2+ ions also. The connections were made. Here the reactor is irradiated with solar light. Reaction mixture is continuously stirred by a magnetic stirrer. The reaction mixture had H_2O_2 dosage 57.6 % , the current density 140.5 A/m^2 , reaction time 45 min. and pH 2.9. After 45 minutes treated samples were allowed to settle for 2 hours and the supernatant was used for analysing the parameters. The experiment was also done with the original leachate.

III. RESULTS AND DISCUSSIONS

A. Treatment of landfill leachate by Electro Fenton Process

Table 2 demonstrates the design of experiments according to Box Behnken design and responses, for example, the removal of COD, Ammonia nitrogen, Chloride, Sulfate, Sulfide, and Phosphate. A second order polynomial model was fitted to the exploratory data for COD removal as given underneath.

$$Y = b_o + b_1X_1 + b_2X_2 + b_3X_3 + b_4X_4 + b_{11}X_1^2 + b_{22}X_2^2 + b_{33}X_3^2 + b_{44}X_4^2 + b_{12}X_1X_2 + b_{13}X_1X_3 + b_{14}X_1X_4 + b_{23}X_2X_3 + b_{24}X_2X_4 + b_{34}X_3X_4 \quad (10)$$

Where Y is the predicted value, X_1, X_2, X_3, X_4 are the variables, b_o is a constant, b_1, b_2, b_3, b_4 are regression coefficients for linear effects. b_{11}, b_{22}, b_{33}, b_{44} are quadratic coefficients and b_{12}, b_{13}, b_{14}, b_{23}, b_{24}, b_{34} are the interaction coefficients.

For COD removal, the acquired coefficient of determination (R^2) is 0.9506. This infers 95.06% of the variations in percent removal of COD are clarified by the independent variables. Table 3 demonstrates the regression coefficients and a p value of various independent variables for removal of COD. Low p value shows that the corresponding coefficient value is more significant. For this situation, the linear effect and quadratic effect of the factors were huge with the exception of the linear effect of pH. Higher p value for interaction coefficients demonstrates their lesser influence. pH - reaction time and H_2O_2 dosage - reaction time were related and had a huge interactive impact .

Table 2 The design of experiment and experimental response for Electro Fenton treatment

SL. No:	pH	Current density (A/m^2)	Dosage (%)	Time (min)	Pollutant removal (%)					
					COD	Ammonia Nitrogen	Chloride	Sulphate	Sulphide	Phosphate
1	2	15.0	50.0	37.5	74.2	39.1	59.0	56.1	52.0	59.1
2	4	15.0	50.0	37.5	72.0	37.3	57.2	52.9	50.0	57.4
3	2	150.0	50.0	37.5	78.2	41.8	63.1	58.8	56.0	62.4
4	4	150.0	50.0	37.5	77.9	42.0	60.5	59.0	55.0	62.1
5	3	82.5	20.0	15.0	74.2	37.2	59.7	57.6	52.5	57.1
6	3	82.5	80.0	15.0	78.1	39.3	62.9	57.2	54.6	59.3
7	3	82.5	20.0	60.0	78.0	39.0	60.9	56.3	54.0	59.4
8	3	82.5	80.0	60.0	76.3	40.9	61.0	58.1	54.0	61.2
9	2	82.5	50.0	15.0	76.4	40.0	60.7	56.9	53.0	60.1
10	4	82.5	50.0	15.0	73.9	38.3	59.2	57.2	52.0	58.3
11	2	82.5	50.0	60.0	74.8	41.7	60.0	57.3	53.0	62.3
12	4	82.5	50.0	60.0	78.0	40.1	60.2	56.8	54.0	60.0
13	3	15.0	20.0	37.5	73.1	37.2	58.3	52.0	51.0	57.4
14	3	150.0	20.0	37.5	76.5	40.8	61.0	58.8	56.4	61.2
15	3	15.0	80.0	37.5	75.0	38.2	60.0	57.0	52.9	58.1
16	3	150.0	80.0	37.5	77.9	41.0	63.8	59.9	56.1	60.0
17	2	82.5	20.0	37.5	76.0	40.0	60.0	56.9	53.2	59.8
18	4	82.5	20.0	37.5	74.2	37.1	59.4	55.4	51.9	57.4
19	2	82.5	80.0	37.5	76.0	40.0	61.1	59.2	54.0	60.2
20	4	82.5	80.0	37.5	76.2	40.2	59.4	57.1	53.2	60.1
21	3	15.0	50.0	15.0	73.0	37.2	58.2	53.6	51.3	57.3
22	3	150.0	50.0	15.0	78.4	39.9	62.1	59.2	54.0	60.0
23	3	15.0	50.0	60.0	74.0	40.1	59.0	54.0	52.0	60.4
24	3	150.0	50.0	60.0	77.9	42.0	62.5	58.9	57.3	62.3
25	3	82.5	50.0	37.5	77.8	40.9	62.9	60.0	55.9	61.6
26	3	82.5	50.0	37.5	78.0	41.0	62.8	60.0	56.2	61.7
27	3	82.5	50.0	37.5	78.0	41.1	63.0	59.6	56.1	61.8

Fig. 2 demonstrates the optimum estimations of every parameter. The optimum conditions acquired from the Minitab are pH = 2.85, current density = 140.5 A/m^2, H_2O_2 dosage = 53% and reaction time = 44 min.

As indicated by the general standards for release of environmental pollutants into surface water Part-A: Effluents [Schedule VI] according to The Environment (Protection) Rules, 1986, Govt. of India, the COD ought to be under 250 mg/l. The leachate effluent after EF treatment can't be securely released into surface water. BOD to COD ratio increased from

0.21 to 0.43. For effective biodegradation, the effluent must have a BOD to COD ratio (biodegradability index) of not less than 0.4 [15]. This demonstrates EF process oxidizes the refractory natural or inorganic mixes of the landfill leachate in a more proficient way. Matured landfill leachate after EF treatment can be subjected to consequent biological treatment for further degradation.

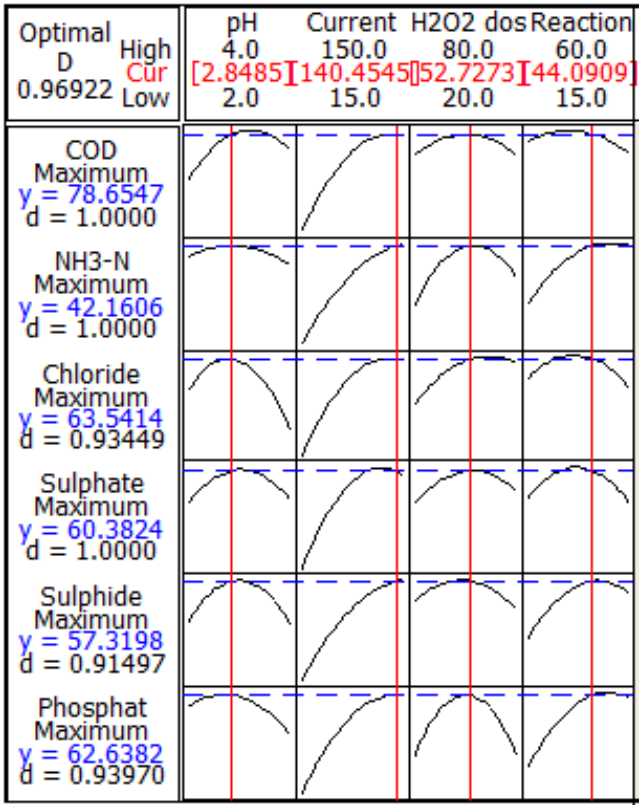

Fig. 2: The graph showing optimum values of the independent variables

Table 3. Regression coefficients and corresponding p values for COD removal

Term	Regression Coefficient	P	Term	Regression Coefficient	P
b_o	62.6564	0.000	b_{44}	-0.0013	0.030
b_1	3.9278	0.151	b_{12}	0.0070	0.164
b_2	0.0696	0.000	b_{13}	0.0167	0.144
b_3	0.1505	0.005	b_{14}	0.0633	0.001
b_4	0.0538	0.044	b_{23}	-0.0001	0.703
b_{11}	-1.3333	0.000	b_{24}	-0.0002	0.264
b_{22}	-0.0003	0.001	b_{34}	-0.0021	0.001
b_{33}	-0.0009	0.008			

Table4 Pollutant removal efficiencies after EF process

Parameter	Concentration before treatment	Concentration after EF process	Removal % after EF process
TSS (mg/l)	498	102	79.5
BOD (mg/l)	4872	2173	58.5
COD (mg/l)	23200	5058	78.2
Ammonia Nitrogen(mg/l)	2196	1296	41.0
Phosphate (mg/l)	185	72.3	60.9
Sulphate (mg/l)	587	240.7	59.0
Sulphide (mg/l)	20.1	8.9	55.8
Chloride (mg/l)	2670	1009	62.2
BOD/COD	0.21	0.43	

B. Treatment of landfill leachate by Photo Electro Fenton Process

EF reactor was irradiated with UV light under optimum conditions. The effluent is analyzed and result is given in table 4. In PEF method H_2O_2 in the presence of Fe^{2+} and UV illumination are used to mineralize the pollutants. The action of this irradiation is complex and can be described by: (a) the increase in the production of hydroxyl radical from photo reduction of $Fe(OH)^{2+}$ and (b) the predominant Fe^{3+} species in acid medium and the photolysis of complexes of Fe(III) with generated carboxylic acids. Biodegradability in terms of BOD/COD ratio of the wastewater improved to 0.36. The pollutant removal efficiencies are shown in table 4.

Table4 Pollutant removal efficiencies after PEF process

Parameter	Concentration before treatment	Concentration after PEF process	Removal % after PEF
TSS (mg/l)	498	72.7	85.4
BOD (mg/l)	4872	1764	63.8
COD (mg/l)	23200	4849	79.1
Ammonia Nitrogen(mg/l)	2196	1099.2	50.1
Phosphate (mg/l)	185	55.1	70
Sulphate (mg/l)	587	187.8	68
Sulphide (mg/l)	20.1	6.2	59
Chloride (mg/l)	2670	961.2	64
BOD/COD	0.21	0.36	

C. Treatment of landfill leachate by Solar Photo Electro Fenton Process

EF reactor was irradiated with solar light under optimum conditions. Biodegradability of landfill leachate improved from 0.21 to 0.38. The pollutant removal efficiencies are shown in table 5.The illumination with UV light in PEF and sunlight in SPEF enhances the degradation process due to the Photochemical process (photolysis) of complexes of Fe(III) with generated carboxylic acids. Graphical representation of comparison between EF, PEF and SPEF are given in fig.3.

Table 5 Pollutant removal efficiencies after SPEF process

Parameter	Concentration before treatment	Concentration after SPEF process	Removal % after SPEF
TSS (mg/l)	498	101.6	79.6
BOD (mg/l)	4872	1910	60.8
COD (mg/l)	23200	5034	78.3
Ammonia Nitrogen(mg/l)	2196	1153	47.5
Phosphate (mg/l)	185	60.1	68.5
Sulphate (mg/l)	587	228.9	61
Sulphide (mg/l)	20.1	7.3	58.6
Chloride (mg/l)	2670	1036	62.4
BOD/COD	0.21	0.38	

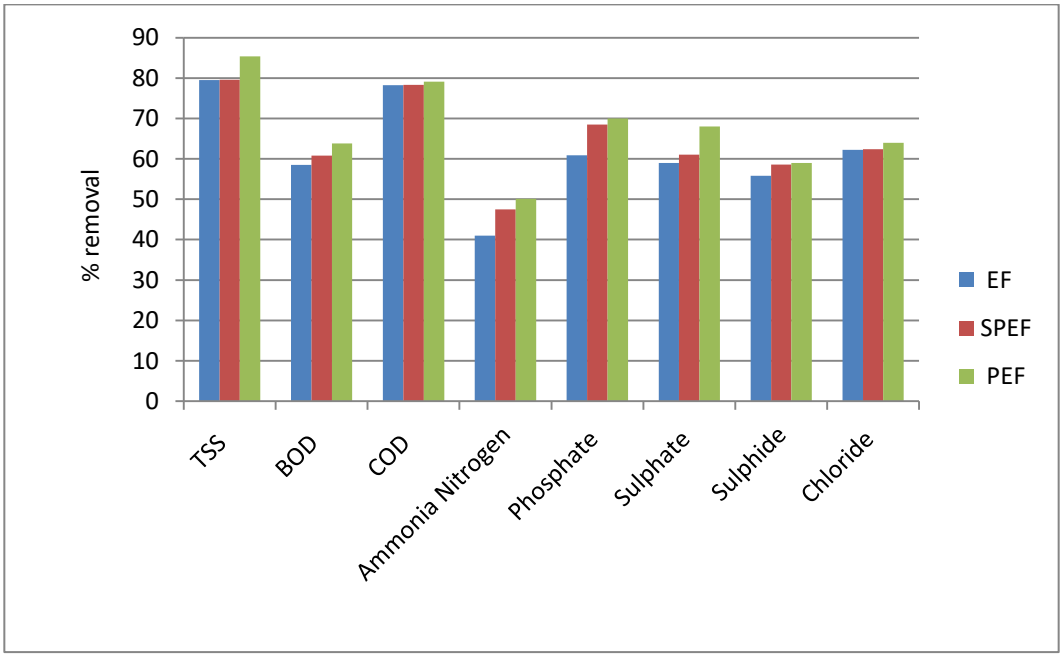

Fig. 3: The percentage degradation of pollutants after EF,PEF and SPEF processes

CONCLUSION

A laboratory examination was done to assess the appropriateness of Electro Fenton (EF) process, Photo electro fenton process using UV and Solar lights for the treatment of a matured municipal landfill leachate gathered from Municipal Solid Waste Disposal Facility at Brahmapuram, Kochi. The impacts of significant parameters on EF procedures and optimization were assessed utilizing Response surface methodology. The ideal conditions for maximum removal of pollutants obtained from the Minitab programming are pH =2.85, current density = 140.5 A/m^2, H_2O_2 dosage = 53% and reaction time = 44 min for EF treatment. After EF treatment, the percentage removal obtained for BOD, COD, ammonia nitrogen, phosphate, sulfate, sulfide, and chloride are 58.5, 78.2, 41.0, 60.9, 59.0, 55.8 and 62.2% respectively. BOD to COD ratio increased from 0.21 to 0.43 which shows that the biodegradability of the waste water has improved. After the AOP treatment the results shows that the AOP treatment alone are not sufficient for the effective treatment of landfill leachate. In light of the important Indian Standards, the treated leachate effluent can't be released into surface water. Irradiating EF reactor with UV light and solar light at optimum conditions enhanced the degradation of pollutants but biodegradability index were less than 0.4. Hence complete biological degradation of pollutants in a biological treatment unit is not possible after PEF or SPEF pretreatment. It can be concluded that the EF process can be utilized as a successful pretreatment technique for matured landfill leachate that is hard to treat with conventional biological treatment technologies.

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