ISSN (ONLINE): 2454-9762
ISSN (PRINT): 2454-9762
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IJARMATE
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International Journal of Advanced Research in Management, Architecture, Technology and Engineering (IJARMATE)
Vol. 3, Special Issue 2, April 2017

A feasibility study of fenton and photo fenton in the treatment of leachate

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Abstract— ***Leachates contain some macromolecular organic substances that are resistant to biological degradation. Recently, Fenton's oxidation has been investigated for chemical treatment or pre-treatment of leachate. Fenton process is one of the powerful and environmentally friendly technologies for treatment of leachate. This article reviews the fundamental and recent developments in fenton process for treatment of leachate. The effects of various operating conditions such as reaction time, initial pH and dosage of hydrogen peroxide, Fe^{2+} ions and their optimum ranges for maximum pollutant removal and mineralization are reviewed. Characterisation of leachate and it effects are also analysed. From this analysis at certain limits of pH the pollutant are removed efficiently.***

Keywords—fenton; leachate; photo fenton; pollutants; treatment; ground water

I. INTRODUCTION

Sanitary landfills are the primary method currently used for municipal solid waste disposal in many countries, and leachate generated from landfills is a high-strength wastewater exhibiting acute and chronic toxicity. Untreated leachates can permeate ground water or mix with surface waters and contribute to the pollution of soil, ground water, and surface water [1]. Urban development, industrial and commercial growth together with increasing population is accompanied by ever increasing solid waste generation all over the world. Due to economic advantages, landfilling is still the most adopted solid waste disposal methods. The inevitable drawback of landfilling is the generation of leachate produced by physico-chemical and biological decomposition of waste and percolation of rainwater through compacted wastes [2], the proper treatment and safe disposal of the leachate is one of the major environmental challenges worldwide especially in Developing countries like India. However, leachate Composition may vary widely within the stages three types of leachates can be defined according to landfill age [3].

The below Table-1 will show the leachate type, year, BOD, COD ratio, TOC/COD ratio, nitrogen content for all three young, intermediate and stabilized leachate type. Mainly two different phases can be identified in landfills during the anaerobic decomposition of waste: acid phase, which causes a decrease of pH in the leachate but high concentrations organic acids and inorganic ions (for example, Cl-, SO42-, Ca2+, Mg2+, Na+) and the methanogenic phase.

TABLE - I

Leachate Type	Young	Intermediate	Stabilized
Landfill Age (Year)	< 5	5 – 10	>10
COD g/l	> 20	3 – 15	< 2
BOD/COD	> 0.3	0.1 – 0.3	< 0.1
TOC/COD	0.3	-	0.4
Nitrogen		100 – 2000mg/l TKN	

Heavy metal concentrations are in general comparatively low. Leachate from the acid phase is therefore characterized by high BOD5 values (commonly > 10.000 mg/l), high BOD5/COD ratios (commonly > 0.7) and acidic pH values (typically 5 - 6). Further information's on the biological degradation processes can be found elsewhere (Stegmann and Spendlin, 1989). The stable methanogenic phase (Phase IV, Fig. 1) of anaerobic degradation is characterized by a pH range from 6 to 8. At this stage, the composition of leachate is characterized by relatively low BOD values and low ratios of BOD/COD. Ammonia continues to stay at a relatively high level. It becomes obvious that the organics (COD, BOD5, TOC) as well as AOX, SO4, Ca, Mg, Fe, Mn, Zn and Cr are highly influenced by the acid phase resp. methanogenic phase. Kruse (1994) investigated 33 landfills in Northern Germany, the leachate concentrations mainly derive from the late eighties and early nineties. He defined three characteristic periods according to the BOD5/COD-ratio:

Acid phase: $BOD5/COD \geq 0.4$

Transient phase: $0.4 > BOD5/COD > 0.2$

Methanogenic phase: $BOD5/COD \leq 0.2$

Between the two investigations there are significant differences concerning the organic parameters. In the younger landfills (Kruse, 1994) leachate concentrations of COD, BOD5 and TOC are lower than those determined by Ehrig (1990) some ten years before. This can be explained by developments in the technology of waste landfilling where in many younger landfills waste compaction is practised in thin layers. In addition also the waste composition may have changed (less

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Available online at www.ijarmate.com

IJARMATE
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(IJARMATE) Vol. 3, Special Issue 2, April 2017

biodegradable waste). These effects may result in a shortening of the acid phase and to an accelerated production of methane and carbon dioxide [4].

To treat these old landfill leachates, many physical/chemical and biological process have been used. Coagulation and flocculation is a relatively simple technique that may be employed successfully in treating old leachate. However ,this treatment only leads to moderate removal of COD and its main drawback is more sludge production and in some cases traditional chemical coagulants are used its increase on the concentration of iron and aluminium in the liquid phase. Therefore coagulation/precipitation is not appropriate full treatment of leachate due to its limited efficiency for removal of pollutant [5]. One available technology widely used to treat landfill leachate in recent years is Fenton's oxidation process (H_2O_2/Fe^{2+}) which has the advantage of both oxidation and coagulation process. The Fenton reaction can be effectively exploited to treat landfill leachate and may be particularly appropriate for mature leachate.

II. FENTON PROCESS

Advanced oxidation process (AOP) such as Fenton has been proposed as an alternative for the treatment of water contaminated with pesticides. This process is characterized by the production of hydroxyl radicals (OH•) by catalytic decomposition of hydrogen peroxide (H2O2) in reaction with ferrous ions (Fe2+) (Eq.1). Additional reactions occur in the presence of light that produce hydroxyl radicals or increase the production rate of hydroxyl radicals as seen in Eq.(2).These radicals are extraordinarily reactive (oxidation potential 2.8 V) and attack most organic molecules. The obtained Fe3+ or its complexes subsequently act as the light absorbing species that produces another radical while the initial Fe^{2+} is regenerated as seen in Eq. (2)

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

$$Fe3+ + H2O2 + hv \rightarrow Fe^{2+} + H^{+} + OH^{\bullet} \ (2)$$

That $OH^{\cdot}$ Radical react with pollutant and it is removed [6].One available technology widely used to treat landfill leachate in recent years is Fenton's oxidation process (H_2O_2/Fe^{2+}) which has the advantage of both oxidation and coagulation process. The fenton reaction can be effectively exploited to treat landfill leachate and may be particularly appropriate for mature leachate .the relative importance of oxidation and coagulation depends primarily on the (H_2O_2/Fe^{2+}) ratio. Chemical coagulation predominant at slower (H_2O_2/Fe^{2+}) ratio, whereas chemical oxidation dominant at higher (H_2O_2/Fe^{2+}) ratio. Generally, initial P_H 2- 4.5 favours the Fenton reaction [5].

III. TYPES OF FENTON PROCESS

- Homogenous Fenton process
- Heterogeneous Fenton process
- Photo Fenton process

In homogenous Fenton process Fe^{2+} ions are added in liquid phase but it`s main drawback is separate of ions is difficult. In heterogeneous process Fe^{2+} ions are added in solid phase so it separation is easy. Fe^{2+} is easily transfer to Fe^{3+} so it take less time to oxidize the pollutant. In photo Fenton process light sources are used to achieve high efficient of removal of pollutant. In mostly UV rays are used in Fenton process, it`s efficiency is more but UV are passed is high cost. So we have to plan to use natural energy (i.e.) sunlight. Earth receives 1.7x1014 kWh, meaning 1.5x1018 kWh per year of solar radiation, which is approximately; 28000 time the world energy consumption per year. In a tropical country like India ((8º4"-37º6" N latitude), highest level of global solar UV radiation is received.

Adequate amount of Solar UV radiation is received for almost 10 months a year. Average mean peak irradiance of Solar UV- A is 47 W/m2 - 66 W/m2 and average mean peak irradiance of Solar UV- B is 0.195 W/m2 - 0.3384 W/m2 [32]. Nearly, 95- 98% of the sun ultraviolet radiation reaching the earth's surface is UV- A. Only 2-5% of UV light at the earth surface is solar UV - B. Practically all of UV - C and much of UV- B is absorbed by the ozone and the atmosphere. [7]

IV. FACTORS AFFECTING FENTON PROCESS

A. *pH*

The PH is one of the most important factors for Fenton's process. The most suitable range for fenton processes is PH 3. This is due to the higher H2O2 productions. For solution at PH more than 3.5 this equilibrium is controlled by Fe^{2+} ions, under these conditions the removal of pollutant is removed by sorption and not by fenton process. If PH<3.5 fenton process remove the pollutant. At low PH <2, H_2O_2 react with Fe^{2+} is very slowly. [14]

B. *Concentration of H_2O_2*

H_2O_2 plays the role of an oxidizing agent in fenton process. In generally H_2O_2 concentrations increases pollutant removal until optimum H_2O_2 concentrations. This is due to higher concentrations of H_2O_2 produce more hydroxyl radicals to complete mineralisation of pollutant. but if add more concentration of H_2O_2 leads to decrease of removal of pollutant due to scavenger of highly potent OH radicals are generated so initial control H_2O_2 concentration are very important.[14].

C. *Catalyst Concentration*

Catalyst is an important parameter affecting the efficiency of all fenton process. Generally pollutant removal is directly directionally proportional to concentration of catalyst. This is mainly due to increase in active sites to produce OH radical it's to degrade the pollutant. In higher rate of catalyst, to form higher rate of cavities formation this leads to reduce OH radical in the solution and thus reduce the removal efficiency of the system. [14]

D. Temperature

Temperature is one of the important factor influencing the Fenton process. Too low and too high temperature negatively

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Available online at www.ijarmate.com

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impact the process efficiency. The increase in temperature, increase of the reaction of the OH formation. If temperature is too high, decrease of efficiency of Fenton process due to thermal decomposition of the hydrogen peroxide resulted in the decrease of formation of hydroxyl radical. [14]

V. STUDY AREA

Ariyamangalam is situated along the national highway of Trichy to Thanjavur. In earlier it is outer area of corporation of Tiruchirappalli due to urbanization and expansion of the city now it is placed in centre area of Trichy. It is 10 km in the east direction from Trichy. It has an area of 48 hectares. On an average of 20 trucks used to collect the solid waste in and around Trichy Corporation and dumped at ariyamangalam composting yard. There is no proper segregation and disposal of municipal solid waste. Now one manure form unit at ariyamangalam, to segregate biodegradable waste and decompose it to form manure but solid waste generation rate is more. It has been dumped at nearly 20 feet height. Refer Table-II.

TABLE II

S.No	Description	Details
1	Total area (acres)	49
2	Area filled (acres)	47
3	Maximum height of garbage (feet)	20
4	Average height of garbage (feet)	14
5	Current rate of dumping (Tons/day)	200
6	No.of trips to collect to solid waste	60
7	Depth of ground water	21

***Ariyamangalam dumpsite details**

TABLE III

Tests	Control sample -1	Control sample - 2	Sample 1	Sample 2	Sample 3	Sample 4
Sulphate (mg/l)	198.343	179.619	1011.67	85.4	917.85	799.95
Alkalinity	86	78	135	45	110	50
Dissolved oxygen (mg/l)	4.3	5.2	5.8	1.3	5	4.8
Hardness (mg/l)		467	290	255	280	285
pH	7	7.6	6.18	6.24	6.29	6.28
Turbidity	0	0	0	0	0	0
BOD	2.1	2	39	42.7	27	18.8
COD	4	3.7	23	61	64	58
Total dissolved solids	427	459	391	405	399	389

***Water testing parameter**

The control sample one and two are collected around the ariyamagalam area about 10km in north and east side of the study area, Sample one, two, three and four are collected on the dumpsite in regular intervals and the samples are tested in the lab with standard procedure.

Every water testing parameter has desirable limits the values are compared with the characteristics of the sample collected . Desirable limits for sulphate is 150 – 400 mg/l, for alkalinity the desirable limit is upto 100mg/l, dissolved oxygen desirable limit is 4 – 6mg/l, Hardness the limit is 300 – 600mg/l, pH = 6.5 to 8.5, turbidity = 0 to 5 NTU, and for the dissolved solids the desirable limit is upto 500mg/l.

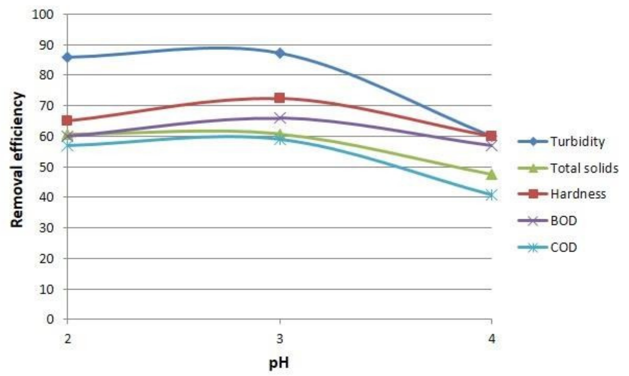
VI. RESULT AND DISSCUSSION

A. FENTON PROCESS

I. Effect of pH

Fig.1. Removal efficiency vs pH

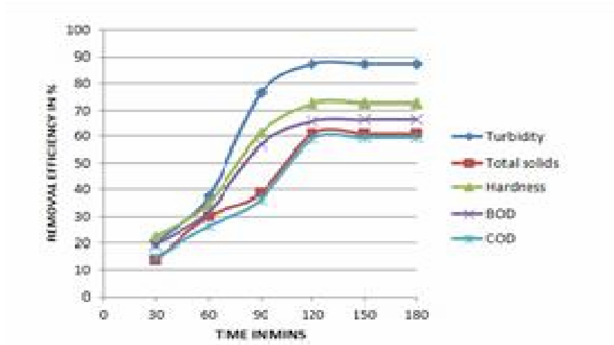
To maintain the ratio is H2O2 /Fe^{2+} = 2 and reaction time is 120min. From the figure it is shown that the maximum



Removal efficiency in turbidity, total solids, hardness, BOD and COD is 87, 60, 72, 65, 59% respectively. This is the effective pollutant removal of 120 min at pH =3.

II. Effect of reaction time

Fig.2. Removal efficiency vs time



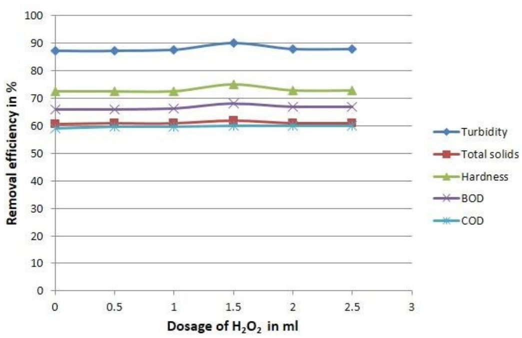
To maintain pH = 3 and the ratio of H_2O_2/Fe^{2+} = 2, From the above figure it is shown that the maximum removal efficiency in turbidity, total solids, hardness, BOD and COD is 87, 60, 72, 65, and 59% respectively this is the effective pollutant removal of 120 min at pH =3, after 120 min the graph will show that there is no change in the removal efficiency therefore we conclude that at 120 min and at pH=3 the removal efficiency of the pollutant is high.

ISSN (ONLINE): 2454-9762
ISSN (PRINT): 2454-9762
Available online at www.ijarmate.com

International Journal of Advanced Research in Management, Architecture, Technology and Engineering
(IJARMATE) Vol. 3, Special Issue 2, April 2017

III. Effect of hydrogenperoxide

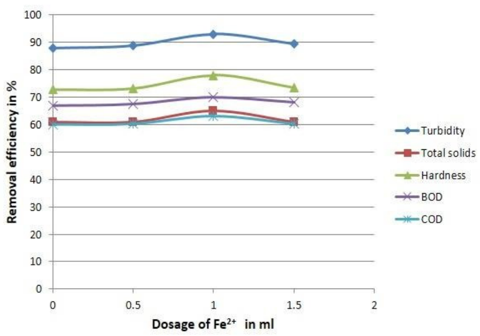
Fig.3. Removal efficiency vs Dosage of H_2O_2



To maintain the pH =3,Reaction time =120 min, ratio of H2O2 /Fe^{2+} = 2. From the fig it is shown that the maximum removal efficiency in turbidity, total solids, hardness, BOD and COD is 90, 62, 75, 68, 60% respectively. This is the effective pollutant removal of 120 min at pH =3, H2O2 /Fe^{2+} = 2 and H2O2 =1.5ml.

IV. Effect of Fe^{2+}

Fig.4. Removal Efficiency vs Dosage of Fe^{2+}



To maintain the pH =3,Reaction time =120 min, ratio of H2O2 /Fe^{2+} = 2 and dosage of H2O2 =1.5ml. From the figure it is shown that the maximum removal efficiency in turbidity, total solids, hardness, BOD and COD is 93, 65, 78, 70, 63% respectively. This is the effective pollutant removal of 120 min at pH =3, H2O2 /Fe^{2+} = 2, H2O2 =1.5ml and Fe^{2+} = 1 ml. Finally from all the tables and graph shows the maximum pollutant removal in Fenton's process and its efficient operating conditions of Fenton's process also shown as above graph.

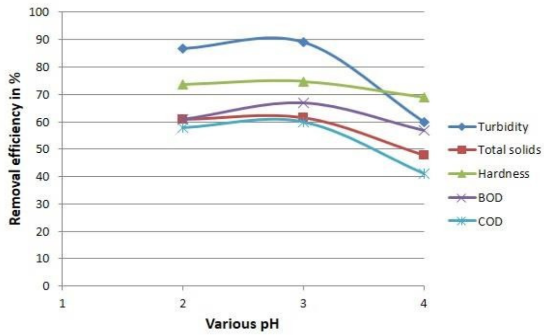
B. PHOTO FENTON PROCESS

UV rays are used in Fenton process, it`s efficiency is more but UV are passed is high cost. So we have to plan to use natural energy (i.e.) sunlight. Earth receives 1.7x1014 kWh, meaning 1.5x1018 kWh per year of solar radiation, which is approximately; 28000 time the world energy consumption per year.

I.

Effect of pH

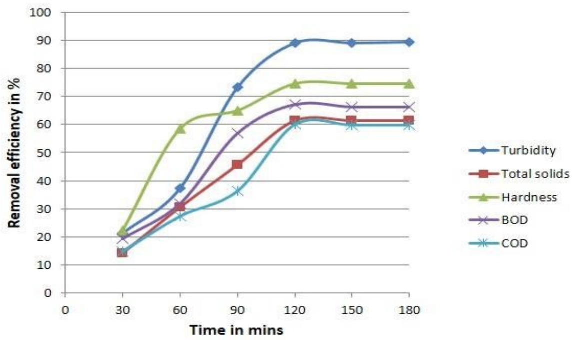
Fig.5. Removal efficiency vs pH



To maintain the ratio is H2O2 /Fe^{2+} = 2 and reaction time is 120min. From the fig it is shown that the maximum removal efficiency in turbidity, total solids, hardness, BOD and COD is 89, 61, 74, 67, 60% respectively. This is the effective pollutant removal of 120 min at pH=3.

II. Effect of reactiontime

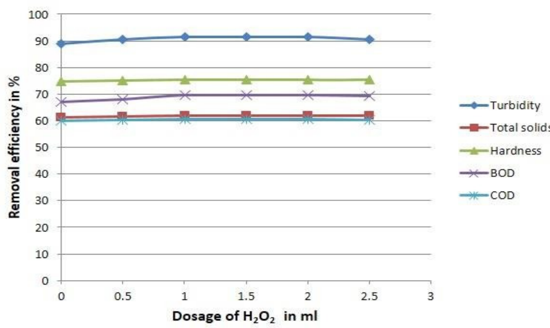
Fig.6. Removal efficiency vs time



To maintain the ratio is H_2O_2 /Fe^{2+} =2 and pH=3, From the fig it is shown that the maximum removal efficiency in turbidity, total solids, hardness, BOD and COD is 89, 61, 74, 67, 60% respectively. This is the effective pollutant removal of 120 min at pH=3.

III. Effect of hydrogenper oxide

Fig.7. Removal efficiency vs Dosage of H_2O_2



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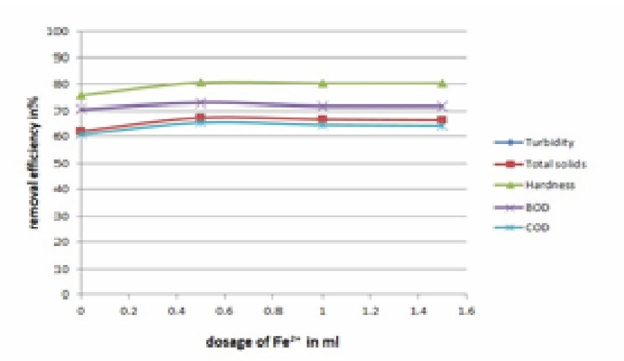
ISSN (PRINT): 2454-9762
Available online at www.ijarmate.com

International Journal of Advanced Research in Management, Architecture, Technology and Engineering (IJARMATE)
Vol. 3, Special Issue 2, April 2017

To maintain the ratio is $H_2O_2/Fe^{2+}=2$, reaction time is 120 min and pH = 3, From the figure it is shown that the maximum removal efficiency in turbidity, total solids, hardness, BOD and COD is 91, 62, 75, 69, 60% respectively. This is the effective pollutant removal of 120 min at pH =3, H2O2 /Fe^{2+} = 2 and H2O2 =1.5ml.

IV. Effect of Fe^{2+}

Fig.4. Removal Efficiency vs Dosage of Fe^{2+}



Volume of sample = 500ml

pH of sample = 3

Ratio of $H_2O_2/Fe^{2+} = 2$

Dosage of H_2O_2 = 1ml

From the figure it is shown that the maximum removal efficiency in turbidity, total solids, hardness, BOD and COD is 94, 68, 80, 73, 65% respectively. This is the effective pollutant removal of 120 min at pH=3, H_2O_2 /Fe^{2+} = 2, H_2O_2 =1ml and Fe^{2+} = 0.5 ml.

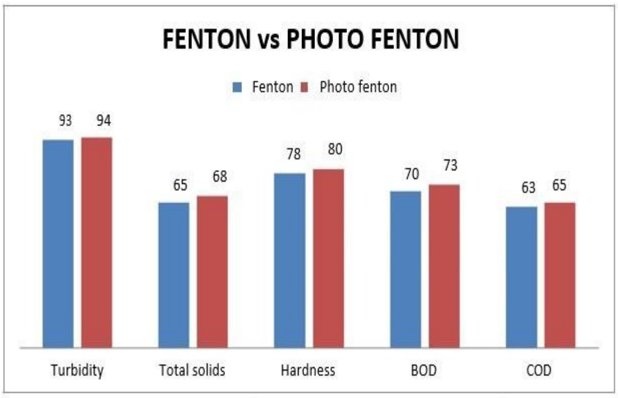
VIII. CONCLUSION

Leachate are one of the major wastewater from solid waste and it threaten to our environment. It percolates below the ground surface and pollute the ground water. Fenton process are one of the powerful tool for treatment of leachate. All these process very much depends on PH, catalyst concentration, H2O2 concentration and temperature.

I. Comparison of fenton and photo fenton process

FENTON		PHOTO FENTON	
Maximum operating condition		Maximum operating condition	
pH	3	pH	3
H2O2 /Fe^{2+}	2	H2O2 /Fe^{2+}	2
Reaction time	120	Reaction time	120
H2O2	1.5	H2O2	1
Fe^{2+}	1	Fe^{2+}	0.5

There exists an optimal value for PH, catalyst concentration, H_2O_2 concentrations and temperature in all the process.



From the above graph, it has shown that the Photo Fenton is slightly more removal efficiency compared to fenton process.

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