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Study of Landfill Leachate Treatment Using Photo-Fenton Process -A Review

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Abstract:- Landfilling is the most common method of solid waste management in our country. Leachate management is a major problem concerned with landfill. Leachate can pollute surface and subsurface water sources. Various studies have been done to obtain effluent of discharge standards. Advanced oxidation processes such as photo-fenton is an effective treatment method in which hydroxyl radicals are generated which help in pollutant degradation. Here, we review conventional treatment methods ,the effect of different parameters such as dosages of hydrogen peroxide, ferrous ion, temperature, initial pH, reaction time and radiation intensity on the efficiency of photo-fenton process. We also summarize different integrated system for leachate treatment using photo-fenton and other processes.

Keywords:- hydroxyl radical, integrated leachate treatment, landfill leachate, photo-fenton.

I. INTRODUCTION

The landfill is the most common technology used to dispose municipal solid residues in developing countries. Surface water that percolates through the landfill and leaches out organic and inorganic constituents from the solid waste is termed as leachate. Landfill leachate production starts at the early stages of the landfill and continues several decades even after landfill closure. Landfill leachate is mainly generated by the infiltrating water which passes through the solid waste fill and facilitates transfer of contaminants from solid phase to liquid phase [1].Leachate may contain large amounts of organic matter (biodegradable, but also refractory to biodegradation), where humic-type constituents make an important group [2], as well as ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts [3]. At the same time, the characteristics of the leachate also vary with regard to its composition and volume, and biodegradable matter present in the leachate against time[4]. The pH of initial landfill is 4.5 - 7 and can go up to 6.6 - 7.5 with passage of time. The BOD will go up to the maximum value when the normal land filling is processing from 6 months to 2 years. The BOD becomes very deliquescent, which is a main characteristic of BOD. Finally, the BOD start to reduce until the landfill is steady should through 6 - 15 years .Effluent COD values are 3,000 – 60,000mg/l [5]. Leachate may also have a high concentration of metals and contain some hazardous organic chemicals [6].

Many different methods are currently in use to treat the landfill leachate. Advanced oxidation process (AOPs) use the hydroxyl radicals for the degradation of pollutants. 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 advantages of both oxidation and coagulation processes [7] . The wastewater treatment by the overall Fenton process consists of four stages, such as oxidation, neutralization, coagulation/ flocculation, and solid-liquid separation. In the Fenton treatment, the oxidation proceeds mainly by hydroxyl radicals and the coagulation by ferric-oxyhydroxides precipitation. Both processes contribute to the reduction of organic constituents [8].

The degradation of organic pollutants by Fenton-type processes can be significantly accelerated in the presence of ultraviolet light irradiation [9]. The organic matter present in the effluents was oxidized, rather than mineralized, higher percentage removals of COD, BOD, and TOC, respectively, having been observed. For this reason, the photo-Fenton process increased BOD/COD ratio, but reduced BOD/TOC ratio. The process is affected by parameters such as Fe^{2+} , H_2O_2 dosage, reaction time, radiation intensity[10]. UV photo-fenton can also be done in the presence of catalysts such as TiO_2 which is found to increase the treatment efficiency of the process [11]. Solar radiation was found more effective than UV in COD removal. The photo reduction of Fe^{3+} by sunlight maintains the concentration of Fe^{2+} in the reactive medium, guaranteeing the efficiency of the photo-Fenton process even in treatments with low initial concentration of ferrous ions [10]. Solar photo-fenton was also studied as pre-treatment method before biological processes such as activated sludge process. The biodegradability of wastewater has enhanced from 0.19 to 0.4 (measured as BOD_3/COD ratio) after 40 min photochemical treatment time . A COD removal of 88% was observed in one hour photochemical treatment time [12]. Different integrated treatment process with fenton and other process have proven to give better results.

II. OTHER CONVENTIONAL METHODS

To treat the landfill leachate, many physical / chemical (chemical oxidation, chemical precipitation, coagulation-flocculation, activated carbon adsorption and membrane processes) and biophysical processes have been used [13]. Coagulation and flocculation is a relatively simple technique that may be employed successfully in treating old landfill leachate. However, this treatment only leads to moderate removals of COD and TOC, and it has its drawbacks: sludge is produced, and in some cases, when traditional chemical coagulants are used, an increase on the concentration of aluminium or iron, in the liquid phase, may be observed [14]. Lime treatment alone provides removal efficiencies of approximately 50% of the organic matter. The complete chemical-physical treatment sequence consisting of lime precipitation/sedimentation/ammonia stripping achieved the removal efficiency of 48%-69% of the organic matter[15]. A study showed that EC is an efficient process for treating stabilized leachate. A 67% COD removal was observed when electro-coagulation was performed for an electrolysis time of 30 min and a current density of 666.6 A/m^2. The conductivity of the sample increased when fly ash C, fly ash F, and bottom ash were introduced into it [16]. Activated carbon adsorption systems have also been used in the treatment of landfill leachate for the removal of dissolved organics, however, they are generally considered as one of the more expensive treatment options and often, must be combined with other treatment technologies to achieve desired results [17]. The most common biological treatment is activated sludge, which is a suspended-growth process that uses aerobic microorganisms to biodegrade organic contaminants in the leachate [18,19]. Biorefractory contaminants, contained mainly in older leachate, are not amenable to conventional biological processes, whereas the high ammonia content might also be inhibitory to activated sludge microorganisms [20].

Table 1 : **Characteristics and Classification of Landfill Leachate Based on Age [21,22]**.

Parameters	Young	Intermediate	Old
Age (years)	< 5	5-10	>10
pH	<6.5	6.5-7.5	>7.5
COD(mg/L)	>10,000	4,000-10,000	<4,000
BOD_5/COD	0.5-1.0	0.1-0.5	<0.1
TOC/COD	<0.3	0.3-0.5	>0.5
Ammonia nitrogen (mg/L)	<400	-	>400
Heavy metals	Low to medium	Low	Low

III. EFFECT OF PARAMETERS

3.1 Effect of H_2O_2 dosage

H_2O_2 dose is a critical variable in the process. A low concentration of H_2O_2 did not generate enough OH• in solution. Addition of H_2O_2 above optimum will lead to decrease in hydroxyl radical concentration due to free radical scavenging by the excess H_2O_2 [23,24]. So there was an optimum H_2O_2 concentration to achieve the maximum percentage of removal although the definition of the concentration range varies for different contaminated wastes [25,26]. Excessive application of hydrogen peroxide generates gas bubbles, which inhibits sludge sedimentation [28,29]and may be detrimental to biological treatments a posteriori [30]. In a study [27], the photo-Fenton treatment was mediated by a $[H_2O_2]/[Fe^{2+}]$ ratio = 114, and after a total irradiation of 90 kJ/L, it achieved 96.5% removal efficiency for TC, 63.0% for COD, and 75.9% for NH^{4+}.

The effect of mode of reagent addition was also studied by [31]. These authors reported that there was an additional increase of about 10% in COD removal when the reagents were added in a continuous mode rather than when all reagents were added simultaneously in batch mode. Similar results were obtained in other studies [32,33].

3.2 Effect of Fe^{2+} dosage

The amount of ferrous ions is one of the primary parameters that influences the Fenton and photo-Fenton processes. In a study [34], it was observed that the extent of degradation increases with increasing initial Fe^{2+} concentration. Particular attention must be paid to Fe^{2+} dosage in order to avoid the following undesired $HO^{\cdot}$ radicals scavenging reaction occurring in the presence of an excess of Fe^{2+} [35] . Naturally, an excessive dosage of iron can contribute to a significant increase in total dissolved solids (TDS) and electrical conductivity in the effluent. Excessive iron salt dosing requires further treatment of the effluent before its discharge to the receiving water [30]. According to [36], the optimum iron concentration in solar photo-fenton depends essentially on the light source, photo reactor configuration and dimensions and the type of wastewater to be treated. In photo-fenton, the presence of light absorbing species such as fulvic acids , nitrates also affect the optimum iron concentration value [37].

3.3 Effect of initial pH and reaction time.

The solution pH plays an important role in the efficiency of the photo-Fenton reaction, since it greatly influences the molar fraction of the iron-water complexes., iron-organic complexes (e.g. oxalic, formic, etc.) and iron-inorganic complexes (e.g. chlorides, sulphates, etc). [38] indicated that pH 2.8 avoids Fe^{3+} precipitation, and the predominant iron-water species in solution is $FeOH^{2+}$, which is the most photoactive ferric ion water complex and can absorb light until 410 nm. [39] explained higher hydroxyl radical product yields in the pH range of 2–4 by a reaction involving the organometallic complex where either hydrogen peroxide is regenerated or reaction rates are increased.

Reaction time above an optimum doesn't give significant increase in pollutant removal efficiency. According to many studies [40,35], optimum reaction time for photo-fenton is 2 h with less sludge production.

3.4 Effect of temperature

In a study, it was observed that there was no significant increase in COD removal when temperature was increased from 25 to 45 °C by the conventional Fenton process [32]. As temperature increased from 13 to 37^0C, COD removal efficiency increased from 90.3% to 94.3%, from 71.4% to 78.5%, and from 66.6% to 76.6% when initial COD was 1000, 2000, and 3000 mg/L, respectively. Higher temperature was beneficial for organic removal, even though the increase of organic removal is relatively small [41]. In solar photo-fenton , the increase of leachate temperature had a positive effect on the reaction rate, increasing 4 times by raising the temperature from 10^0 C to 40 ^{0}C.This is due to production of more hydroxyl radicals resulting from a higher ferric ion reduction rates through thermal reactions [42].

3.5 Effect of radiation intensity

The rate of initiation of photo catalysis and electron-hole formation in the photochemical reaction is strongly dependent on the light intensity[43]. While increasing the intensity of incident light, the probabilities of excitation of electrons as well as the re-excitation of recombined electrons increase. Hence, increase in the degradation is observed with increase in the intensity of incident light [44]. In some cases, the reaction rate exhibited a square root dependency on the light intensity; others observed a linear relationship between the two variables[45,46]. In a study [47],the optimum radiation required was 57.4 kJ/L of solar UV energy

IV. INTEGRATED LEACHATE TREATMENT WITH PHOTO-FENTON

In photo-fenton process, the initial turbidity should be less to make use of the radiation . Also, the effluent should be treated to obtain the discharge standard. An integrated system with photo-fenton and other processes gives the best results. The treatment of medium-age landfill leachate was investigated by employing several set-ups including a sequencing batch bio-filter granular reactor (SBBGR) step, with ozone enhancement, followed by a polishing stage with solar photo-Fenton [48]. For the target COD 160 mg/L, the combination SBBGR + solar photo-fenton needed 4.1 €/m^3.Another study [49] was done to evaluate the suitability to couple anammox process with advanced oxidation processes (ozonation + photo-fenton) to treat mature landfill leachate with high nitrogen and non-biodegradable organic matter concentrations. Total COD removal efficiency of 98% and total nitrogen removal efficiency of 89%was obtained. Also, physical-chemical treatment followed by solar photo-fenton and then biological treatment enhances the biodegradability of leachate [50,52,59].Post treatment of aerated landfill leachate with fenton also improved the COD and color removal efficiencies rather than increasing the fenton reagent's dosage[51]. Similarly in a study [53], multistage treatment of leachate combining biological nitrification-denitrification / solar photo-fenton / biological process gave effluent of discharge standard.

Table 2: Integrated leachate treatment with corresponding treatment efficiencies

Treatment combination	Parameter with removal %	References	
Aerated lagoon + Photo-fenton + ASP	DOC - 90 % Nitrogen -100%	[54]	
SBBGR + solar photo-fenton	COD -95%	[48]	
Annamox + photo-fenton	COD - 98%	[49]	
Aeration +Fenton + Biological	COD - 89%	[51]	
Electrocoagulation + solar photo-fenton +Biological	COD - 90%	[55]	
Solar photo-fenton + Biological oxidation	Nitrogen - 100%	[56], [58]	
Aerated lagoon + photo-fenton + ASP	DOC - 75% Polyphenol 82%	[57]	

V. CONCLUSION

Landfill leachate management is a major issue which has to be handled properly. Leachate is heavily polluted and thus detrimental to surface and subsurface sources. In the recent years, with the continuous hardening of the discharge standards in most countries and the ageing of landfill sites with more and more stabilized leachate, conventional treatments (biological or physic-chemical) are not sufficient anymore to reach the level of purification needed to fully reduce the negative impact of landfill leachate on the environment.

Photo-fenton process is an efficient method for the degradation of biodegradable and non-biodegradable organic matter in the landfill leachate. Various parameters such as reagent dosage, initial pH, reaction time, temperature, radiation intensity affects the process. Reagent dosage above optimum reduces the efficiency and produces more sludge. An acidic pH range of 2-4 is optimum for photo-fenton process. Temperature increase doesn't have much impact on conventional fenton process ,but in photo-fenton it has a positive impact.

Turbidity affects the photo-fenton efficiency. Thus, pretreatment increases the efficiency .Also, post-treatment of photo-fenton treated leachate by biological process gives better results. Integrated system using physic-chemical, photo-fenton and biological process gives effluent of discharge standards**.**

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