

A comprehensive review of the Fenton-based approaches focusing on landfill leachate treatment

Mujtaba Hussain¹, Mohd Salim Mahtab^{*2} and Izharul Haq Farooqi²

¹Department of Environmental and Management Studies, Al-Falah University, Faridabad, India

²Department of Civil Engineering, Z. H. College of Engineering and Technology, Aligarh Muslim University, Aligarh-202002, Uttar Pradesh, India

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Abstract. Landfilling is the most commonly adopted method for a large quantity of waste disposal. But, the main concern related to landfills is the generation of leachate. The leachate is high strength wastewater that is usually characterized by the presence of high molecular recalcitrant organics. Several conventional methods are adopted for leachate treatment. However, these methods are only suitable for young leachate, having high biodegradability and low toxicity levels. The mature and stabilized leachate needs advanced technologies for its effective treatment. Advanced oxidation processes (AOPs) are very suitable for such complex wastewater treatment as reported in the literature. After going through the literature survey, it can be concluded that Fenton-based approaches are effective for the treatment of various high/low strength wastewaters treatment. The applications of the Fenton-based approaches are widely adopted and well recognized due to their simplicity, cost-effectiveness, and reliability for the reduction of high chemical oxygen demand (COD) as reported in several studies. Besides, the process is relatively economical due to fewer chemical, non-sophisticated instruments, and low energy requirements. In this review, the conventional and advanced Fenton's approaches are explained with their detailed reaction mechanisms and applications for landfill leachate treatment. The effect of influencing factors like pH, the dosage of chemicals, nature of reaction matrix, and reagent ratio on the treatment efficiencies are also emphasized. Furthermore, the discussion regarding the reduction of chemical oxygen demand (COD) and color, increase in biodegradability, removal of humic acids from leachate, combined processes, and the pre/post-treatment options are highlighted. The scope of future studies is summarized to attain sustainable solutions for restrictions associated with these methods for effective leachate treatment.

Keywords: advanced oxidation processes; fenton process, hydroxyl radicals; landfill; leachate treatment

1. Introduction

The increasing population accompanies urbanization and rapid industrialization that leads to large municipal solid waste generation. The management and disposal of this waste is an important concern. In India, more than 90% of the municipal solid waste is finally disposed of on land in a much-unplanned manner (Dasgupta *et al.* 2013). Sanitary landfilling is the widely used practice for

*Corresponding author, Ph.D. Student, E-mail: msmahtab@zhcet.ac.in

the disposal of municipal solid waste. The major distress related to the use of landfills involves the generation of landfill leachate that affects the surrounding environment in many ways. Leachate produced from landfills is high-strength wastewater with varying toxicity levels (Guo *et al.* 2010). The characteristics and composition of landfill leachate largely depend upon the properties of soil, type of solid waste, rainfall patterns, and age of landfill site (Di Iaconi *et al.* 2006). Young landfill leachate (age < 5 years) are considered to be more biodegradable with a presence of low to medium concentration of heavy metals while the intermediate (5-10 years) and old (> 10 years) landfill leachates are characterized by the presence of humic and fulvic acids and other recalcitrant high molecular compounds with very high chemical oxygen demand (COD) values (Renou *et al.* 2008, Mahtab *et al.* 2020). Hence, the age of landfills is an important factor that determines the composition of landfill leachates.

Leachate is also recognized as the source of more than 100 chemicals that contaminate the groundwater (Schwarzbauer *et al.* 2002, Praveen and Sunil 2016, Khan *et al.* 2021a). Major contaminants in the landfill leachate include the presence of heavy metals, ammonia, dissolved organic matter, and xenobiotic organic compounds (Kjeldsen *et al.* 2002, Khan *et al.* 2021b). Proper treatment and disposal of landfill leachates are very important to reduce its adverse effects on the environment and human health.

Numerous methods have been adopted for the treatment of landfill leachate. Biological processes among such methods are the preferable choice considering their simplicity, cost-effectiveness, and reliability for the reduction of high biochemical oxygen demand (BOD) concentrations from the leachates (Hamza *et al.* 2016). Various aerobic biological methods like activated sludge process, rotating biological contractors, oxidation lagoons, sequence batch reactors, biological aerated filters, and extended aerated systems, etc, are the widely established technologies used for the treatment of landfill leachates (Rathnayake and Herath 2018). Biological methods are considered suitable for the treatment of young leachates with a high biodegradability (Bandala *et al.* 2013, Husain *et al.* 2020). The process comes out to be very inefficient in terms of the removal of high molecular recalcitrant organics present in the mature or stabilized leachate (Deng and Englehardt 2007). The conventional biological treatments face the challenges of temperature control, seasonal conditions, large territorial use, and discharge regulations (Deng 2009). The presence of various toxic and recalcitrant pollutants in landfill leachate limits the conventional wastewater treatment schemes and requires other alternative technologies that can efficiently eliminate these pollutants. Several studies conducted for the treatment of landfill leachate systems suggested the advanced oxidation processes (AOPs) for efficient removal of a large spectrum of refractory contaminants present in the leachate (Deng 2009, Sharma *et al.* 2011, Hussain *et al.* 2020, Mahtab *et al.* 2020).

AOPs are the physical-chemical treatment processes that work on the same chemical principle of generation of highly reactive radical species to achieve the degradation of a wide range of organic pollutants present in the water and the wastewater (Gogate and Pandit 2004, Ataei *et al.* 2015, Da Costa *et al.* 2018, Hussain *et al.* 2020). In AOPs, the high oxidation potential of hydroxyl radicals degrade the pollutants and oxidizes them into the simpler intermediates, ultimately, to the carbon dioxide and water (Neyens and Baeyens 2003, Mahtab *et al.* 2020). Some of the commonly used AOPs include the Fenton process, ozonation, hydrogen peroxide oxidation, peroxidation, photocatalysis, ultraviolet irradiation, microwave enhanced AOP, wet air oxidation, and electrochemical oxidation, etc. AOPs can be used as the pre-treatment and post-treatment schemes as well to complement the biological treatments with improved biodegradability and the enhanced removal efficiency of organics (Deng 2009, Raji and Palanivelu 2016, Verma and Haritash 2020). Among the AOPs, the Fenton process is considered to be a very attractive choice

for the effective treatment of recalcitrant organic compounds present in the leachate and other wastewaters (Zazouli *et al.* 2012, Sharma *et al.* 2016, Mahtab *et al.* 2020). The Fenton process is considered to be an effective alternative to oxidize the recalcitrant compounds and convert them into more readily biodegradable intermediates (Ismail and Tawfik 2016).

In this review, the fundamental mechanisms and applications of various Fenton-based approaches were explained focusing on landfill leachate treatment. The effect of influencing factors on the processes' efficacies is also emphasized. Additionally, the discussion about very important topics related to these methods and procedures was covered and the scope of future research studies was also recommended. Although several review articles have been published on related topics. However, the papers focusing on the overview of Fenton-based approaches for landfill leachate treatment explicitly are lacking. This review covered extensive applications of Fenton-based approaches, which will be helpful to the readers working a research on wastewater treatment particularly focusing on AOPs for landfill leachate treatment options.

2. Fenton process

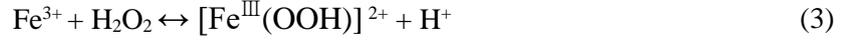
This method was discovered by the British chemist H. J. H. Fenton in the year 1894. The Fenton reaction involves the ferrous ions (Fe^{2+}) catalysts for the active decomposition of hydrogen peroxide (H_2O_2) in an acidic condition to produce the highly reactive hydroxyl radicals (OH^\bullet) (Da Costa *et al.* 2018). The hydroxyl radicals are responsible for the degradation of a large number of recalcitrant and toxic organic compounds present in the water and the wastewater (Leifeld *et al.* 2018). Simultaneous oxidation and flocculation in the Fenton process result in the improved removal of organics (Barbusiński and Filipek 2000). The operational parameters are pH, reaction time, temperature, H_2O_2 , and Fe^{2+} ratio (Fenton's reagent ratio) and organic pollutants concentration, etc. Fenton's reagent ratio and wastewater pH are the crucial factors that greatly affect the treatment efficiency of the Fenton process (Bello *et al.* 2019). This process can be more effective in combination with other processes. In general, the process is very effective and leads to the complete mineralization of various organic compounds, however, the immense quantity of chemicals required makes the process very uneconomical for large-scale treatments (Huston and Pignatello 1999).

2.1 Mechanism of the Fenton process

The basic mechanism in the Fenton process involves the reaction between the Fenton's reagents (Fe^{2+} and H_2O_2) for the production of highly reactive hydroxyl radicals ($\bullet\text{OH}$) described as follows



The reaction of Fe^{3+} (ferric ion) with hydrogen peroxide in a Fenton-like reaction leads to the regeneration of Fe^{2+} ions and also produces hydroperoxyl radical (HO_2^\bullet) as shown in Eq. (2). HO_2^\bullet plays an active role in the degradation of a large number of organic pollutants, but it is considered less reactive than the $\bullet\text{OH}$. The rate constant of the Fenton-like reaction is considered to be many orders less than that of Eq. (1) and excess of Fe^{3+} causes the issues of iron sludge production (Deng and Zhao 2015).



A reaction shown in Eq. (2) is associated with the generation of $[\text{Fe}^{\text{III}}(\text{OOH})]^{2+}$ complex, as shown in Eq. (3) that follows the equilibrium principle. They produced complex breaks and leads to the generation of the Fe^{2+} and hydroperoxyl radical (HO^{\bullet}_2) as shown in Eq. (4) (De Laat *et al.* 1999). Fe^{2+} can also be regenerated in the presence of hydroperoxyl radicals (HO^{\bullet}_2), organic radicals (R^{\bullet}), and superoxide ion ($\text{O}_2^{\bullet-}$) through the pathway of the reduction of ferric ions as shown in Eqs. (5)-(7) (Rothschild and Allen 1958, De Heredia *et al.* 2001).



The required dosage of H_2O_2 was based on the initial concentration of pollutants present. The theoretical stoichiometric ratio of H_2O_2 and COD gives the idea of the required dosage of hydrogen peroxide (Lücking *et al.* 1998). Excess of Fe^{2+} and H_2O_2 causes scavenging of hydroxyl radicals as shown in Eqs. (8) and (9). Hence, the experimentally designed optimum molar ratio of Fe^{2+} and H_2O_2 is evaluated to minimize the scavenging effect of hydroxyl radical (Deng and Zhao 2015). Hydroxyl radicals may further be scavenged by the presence of hydroperoxyl radicals and auto scavenging effects as shown in Eqs. (10) and (11) (Babuponnusami and Muthukumar 2014).



Scavenging reactions consumed most of the hydroxyl radicals that in turn lead to the reduced efficiency of the process and make the process uneconomical. Various other reactive radicals like hydroperoxyl radicals and superoxide ions also face scavenging issues as shown in Eqs. (12)-(15) (Stuglik and Pawełzagórski 1981, Bielski *et al.* 1985).



The hydroxyl radicals degrade the organic compounds by the following three types of mechanisms i.e. hydrogen abstraction, hydroxyl addition, and electron transfer (Huang *et al.* 1993). Hydrogen abstraction mechanism involves the hydrogen abstraction reactions of unsaturated organic compounds as shown in Eq. (16) whereas hydroxyl addition mechanism involves the

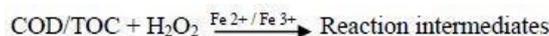
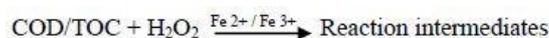
direct hydroxyl radical's reaction with carbon-carbon multiple bonds or aromatic system of organic compounds Eq. (17). Electron transfer mechanisms are usually associated with electron transfer reactions of inorganic ions as shown in Eq. (18) (Bello *et al.* 2019).



The reaction between hydroxyl radicals and organic compounds leads to the formation of carbon-centered radicals (R \cdot), which reacts with the available oxygen in the water and produces complexes and radicals as shown in Eqs. (19) and (20). The coupling of the produced radicals (R \cdot , R-OO \cdot , and R-O \cdot) results in the formation of relatively stable intermediate molecules. The radicals may also react with the iron ions (Pignatello *et al.* 2006). These intermediates further react with oxygen and hydroxyl radicals and eventually get mineralize into the water and carbon dioxide hence completes the mechanism.



In general, the Fenton process can be easily understood by the following reactions (Zazo *et al.* 2005).



2.2 Influencing factors

The treatment efficiencies of the process are influenced by the various operational parameters, Fenton's reagent ratio and pH are the major factors that determine the oxidation capability of the process.

2.2.1 pH

pH is a very important factor that governs the hydroxyl radicals generation in the process. pH range of 2.8-3.5 is considered most favorable (Pouan *et al.* 2015, Villegas-Guzman *et al.* 2017a,b). Too much acidic and alkali condition makes the process ineffective and uneconomical. Extreme lower pH values lead to the formation of [Fe (H₂O)₆]²⁺ species which slows down the process and results in the low yielding of hydroxyl radicals (Xu *et al.* 2009). The high concentration of H₂O₂ present at the low pH values acts as a scavenger of OH \cdot radicals, which causes the formation of oxonium ions [H₃O₂]⁺ ultimately reduced the treatment efficiency of the process (Kavitha and Palanivelu 2005). On the other hand, at pH higher than optimum value, the ferric ions (Fe³⁺) start to precipitate out as ferric hydroxide which inhibits the production of OH \cdot radicals and regeneration of Fe²⁺. Additionally, the higher pH value causes auto decomposition of

H₂O₂ into the carbon dioxide and water that adversely affects the process (Szpyrkowicz *et al.* 2001). With the increase in pH values, the oxidation potential of hydroxyl radicals decreases. The oxidation potential of hydroxyl radical at pH 14 comes out to be 1.95V as compared to the 2.8V at zero pH (Kim and Vogelpohl 1998). The higher pH value limits the decomposition of H₂O₂ into hydroxyl radicals. Also, alkaline and neutral condition favors the occurrence of carbonates and bicarbonates, which are well known for the scavenging of hydroxyl radicals (Gallard *et al.* 1998).

A study performed by Kang and Hwang (2000) evaluated the coagulation efficiency of leachate after Fenton's oxidation over the pH range of 2 to 9. It was concluded that the pH range of 3 to 6 was effective and lead to the highest COD and color removal. However, the conventional Fenton process can be made effective at higher pH values by the incorporation of chelating agents and heterogenous catalysts into the matrix. The chelating agents with their various coordinating sites are well known for their bonding with the metal ions. The addition of the chelating agents in the Fenton process results in the formation of Fe(II)/ Fe(III) complexes that prevent the precipitation of Fe salts and keeps them soluble at higher pH values (Lipczynska and Kochan 2008, Sun and Pignatello 1992). However, the applicable range of pH for the chelate modified Fenton process is determined by the corresponding chelating agent and the iron complexes that are formed by these agents. Various studies reported the applicability of the chelate modified Fenton Process (De Laat *et al.* 2011, Li *et al.* 2001(a) and Sun and Pignatello 1992). In comparison with the other chelating agents, the polyacrylic acids are considered to be very advantageous polymeric chelates due to their multiple binding sites (Li *et al.* 2005). The polyacrylic leachates combine with the Fe(II) and Fe(III) salts at multiple sites and improve their solubility for effective oxidation at higher pH values (Li *et al.* 2007). The use of heterogeneous catalysts such as transition metals, iron oxides, composites, etc is another possible solution to overcome the pH limitation concern in the Fenton process addition of various other species like amino polycarboxylates, polycarboxylates, tartrate, oxalate, and citrate have shown to justify the extension of the applicability of conventional Fenton treatment at neutral or near to neutral pH values (Clarizia *et al.* 2017). A study conducted by Gao *et al.* (2016) employed the heterogeneous Fenton catalyst (i.e. chitosan cross-linked ferrous complex) for the removal of methylene blue and achieved excellent results at a wide range of pH varying from 3 to 7.

2.2.2. Concentration of H₂O₂

Out of the two reagents used in the Fenton process, H₂O₂ is considered to be more important because it determines the overall produced mass of OH[•]. Higher dosage of H₂O₂ results in the higher degradation of pollutants but up to a certain extent. Excess of H₂O₂ causes the scavenging of OH[•] and produced less reactive HO₂[•] that hinders the performance of the process as shown in Eq. (21) (Ahmadi *et al.* 2015).



An excessive dosage of H₂O₂ also lowers down the levels of Fe²⁺ and affects the process with the presence of comparatively more Fe³⁺ ions. The catalytic action of Fe³⁺ is considered many times slower than the Fe²⁺, which in turn reduces the production of OH[•] radicals (Biglarijoo *et al.* 2016). The portion of H₂O₂ that remains unused in the process leads to the increased COD levels in the effluent (Lin and Lo 1997) and hence the dosing of H₂O₂ should be appropriate. Auto decomposition of excess H₂O₂ causes the off-gassing of oxygen that leads to the problem of floating sludge. Further, when the Fenton process is used as a pretreatment, the high concentrations of H₂O₂ negatively impact the microorganisms involved in the associated biological processes (Kim *et al.* 2001, Lau *et al.* 2001). Stepwise addition is an effective way to cease the

increase of H_2O_2 to prevent the rapid decomposition and side reactions of H_2O_2 (Wang *et al.* 2016). The dosage of H_2O_2 is largely dependent upon the initial concentration of COD, high initial COD requires a high dosage of H_2O_2 . The theoretical mass ratio of COD and H_2O_2 is 470.6/1000 (Kang and Hwang 2000).

2.2.3. Concentration of Fe^{2+}

Fe^{2+} acts as the catalyst for the decomposition of H_2O_2 , the higher concentrations of Fe^{2+} result in the increased levels of the OH^\bullet radical's generation. Higher concentrations of Fe^{2+} lead to the higher degradation of pollutants but up to a certain extent. The excess of Fe^{2+} causes the scavenging of OH^\bullet radicals hence affects the efficiency of the process as shown in Eq. (22) (Muangthai *et al.* 2010).



The higher concentration of Fe^{2+} leads to increased total dissolved solids (TDS) levels and the high electrical conductivity of the effluent. It also causes the problem of excessive sludge generation at the end of the process (Gogate and Pandit 2004). The concentration of Fe^{2+} depends upon the dosage of H_2O_2 which in turn depends upon the initial concentration of pollutants.

2.2.4. Reagents ratio ($\text{Fe}^{2+}/\text{H}_2\text{O}_2$)

The concentration of Fe^{2+} and the dosage of H_2O_2 are considered to be very important for the overall efficiency of the Fenton process. The ratio of the reagents (i.e. $\text{Fe}^{2+}/\text{H}_2\text{O}_2$) plays a very important role to determine the optimum required concentrations of OH^\bullet and prevents its scavenging. The iron dosage determines the reaction rate of the process while the extent of mineralization of pollutants largely depends upon the concentration of H_2O_2 . The high ratio of $\text{Fe}^{2+}/\text{H}_2\text{O}_2$ enhances the catalytic decomposition of H_2O_2 and leads to the large generation of OH^\bullet radicals. However, the excess of radicals faces the scavenging effects as shown in Eqs. (8)-(11) and affects the mineralization capacity. Moreover, the high unused concentration of H_2O_2 at the lower $\text{Fe}^{2+}/\text{H}_2\text{O}_2$ ratio also causes the scavenging of OH^\bullet radicals and affects the process efficiency (Gogate and Pandit 2004). Hence the optimum ratio of these reagents is a must to achieve the required treatment efficiency of the process. The optimization of the $\text{Fe}^{2+}/\text{H}_2\text{O}_2$ ratio is usually based on the iteration study performed at the lab-scale. The process of coupling the different dosages of Fe^{2+} and H_2O_2 and selecting the optimal dosage in terms of the highest COD removal is considered to be an effective way to determine the ratio of the optimal reagents (Pala and Erden 2004). Besides, the higher dosage of reagents makes the process uneconomical.

2.2.5. Nature of the reaction matrix

The nature of the reaction matrix is considered to be another important factor that influences Fenton's oxidation. The presence of various organic and inorganic compounds in the wastewater causes the scavenging of OH^\bullet that in turn decreases the efficiency of the process. In the actual wastewater treatment, the presence of various inorganic compounds and mineral chemicals like sodium carbonate, sodium sulfite, and sodium chloride causes the scavenging of OH^\bullet and reduced its availability (Manenti *et al.* 2015). Some of the other well-known scavengers in the wastewater may include natural organic matter (mainly humic acids), bicarbonates, nitrates, etc. The presence of bicarbonates in the wastewater leads to the scavenging of OH^\bullet radicals. The natural organic matter present in the wastewater consumes the large concentrations of OH^\bullet radicals and hence decreases their availability for the oxidation of target organic compounds (Yang *et al.* 2015). The presence of nitrates in the wastewater also results in the scavenging of hydroxyl radicals (Drtinova

et al. 2010). A study conducted by Ghaneian *et al.* (2013) shows that on increasing the concentration of nitrate from 0 to 100 mg/l, the reduction percentage of humic acids has fallen from 51% to 15%. The radical scavenging of OH[•] by nitrate ions is considered to be the main reason for the reduced efficiency.

3. Fenton-based approaches

The various advanced Fenton's approaches are as follows

3.1 Electro-Fenton process

The process involves the use of electrons to complement the conventional Fenton process (CFP). The Electro-Fenton process (EFP) works on the principle of cathodic reduction of Fe³⁺ and O₂ for the generation of Fenton's reagents i.e. Fe²⁺ and H₂O₂ (He and Zhou 2017). Based on Fenton's reagent formation, the EFP can be classified into four types. Type 1 involves the use of oxygen sparging cathode and sacrificial anode for the generation of H₂O₂ and Fe²⁺ respectively with no external addition of reagents (Ting *et al.* 2008). In Type 2, Fe²⁺ is generated from the sacrificial anode while H₂O₂ is externally added. In Type 3, oxygen sparging cathodes are used for the electro-generation of H₂O₂, and Fe²⁺ is externally added (Bello *et al.* 2019). Type 4 involves the electrolytic regeneration of Fe²⁺ by the cathodic reduction of Fe³⁺ ions (Zhang *et al.* 2006). However, type 3 is the most popular EFP that is used for the continuous electro generation of H₂O₂. In a typical process, a continuous supply of oxygen gas at the cathode in an acidic medium causes its two-electron reduction and leads to the formation of H₂O₂ as shown in Eq. (23) (Pliego *et al.* 2015). Initially, a small quantity of ferrous salts is added to the cell to react with H₂O₂ and to generate the Fe³⁺, which in turn continues the cathodic electro regeneration of Fe²⁺ as shown in Eq. (24) (Brillas *et al.* 2009). The sacrificial anode oxidation of iron is also very important in terms of the production of Fe²⁺ as shown in Eq. (25) (Varanket *et al.* 2020).



The process is considered to be very advantageous over the CFP. The electro generation of H₂O₂ could lead to an 80% cost reduction and also saves the associated transport and handling cost. The effective utilization of Fe³⁺ and continuous regeneration of Fe²⁺ minimizes the problem of sludge production and enhances the production of [•]OH (Huang and Chu 2012, Pliego *et al.* 2015). However, several factors like pH, current density, dissolved oxygen level, catalyst concentration, electrolytes, electrode nature, and temperature affect the efficiency of the process. EFP showed the same trend of results for solution pH, temperature, and initial concentration of pollutants as exhibited by the CFP. Applied current is an important factor that determines the electron generation and regeneration of H₂O₂ and Fe²⁺ respectively. The higher applied current leads to higher efficiency but up to a certain limit. The value higher than certain predetermined levels causes the parasitic reactions and adversely affects the performance of the process. Lin and Chang (2000) have reported results in 69% of COD removal and 15.82% of NH₃-N removal for the treatment of landfill leachate by the EFP. The process further increased the biodegradability of

leachate from the value of 0.1 to 0.29. The electrolysis and Fenton process together result in the higher degradation of pollutants as compared to the CFP. In general, the EFP with the high in-situ generation of H₂O₂, electro regeneration of Fe²⁺, and low sludge production is considered to be very advantageous but requires high energy.

The operational costs that are involved in the EFP include labor, material, cost of energy consumption, fixed and disposal costs but the major part of these costs in the EFP comes from the consumption of electric energy (Tirado *et al.* 2018). The high treatment cost due to the high electricity consumption is considered as the main drawback of the EFP. The higher duration of treatment for the effective mineralization of the resistant intermediates formed in the process leads to the higher associated treatment costs (Monteil *et al.* 2019). The higher currents lead to the effective mineralization of contaminants but also add up the higher electric energy consumption. Hence, it is essentially required to properly set the applied current density that marks the balance between the energy-related costs and efficiency of the process (He *et al.* 2017). The consumption of electricity in the electro-Fenton treatment process is analyzed by Eq. (26) given below (Tirado *et al.* 2018).

$$\text{Energy consumption} = \{ U.I.t.1000 / (\text{COD}_0 - \text{COD})V \} \quad (26)$$

where U= Consumed Electric Energy (kWh/kg COD), I = Current Intensity (Amp), T=time (h), V= volume of water (litre), COD₀=Initial COD (mg/l), COD=Final COD (mg/l).

As suggested by several studies, the application of the biological treatment process before the EFP shows the reduction of consumption of electric energy and hence its costs (Monteil *et al.* 2019). A study conducted by Yu *et al.* (2015) shows that the use of modified graphite felt cathode is considered to be more cost-effective for the EFP. A study conducted by Radjenovic and Sedlak (2015) indicates that flows through the three-dimensional electrode configuration may lead to the reduction of energy and costs inputs. Another study claims that the energy consumption costs in the EFP can also be reduced by using alternative sustainable energy sources like MFCs (Microbial Fuel Cells) (De Dios *et al.* 2013).

3.2. Sono-Fenton process

In this process, the high-frequency ultrasound waves are used for the disassociation of water molecules into the hydrogen radical (H[•]) and OH[•] (Eren 2012, Salimi *et al.* 2017). The irradiation of high-frequency ultrasound waves (>15 kHz) causes an adiabatic compression and rarefaction of liquid media. The lower pressure in the rarefaction part of the ultrasonic wave leads to the formation of microbubbles (Gogate 2008). These microbubbles grow and abruptly collapse during the compression phase and create the local high temperature and pressure conditions that may rise to the value of 5000 K and 1000 atm respectively. These short-lived extreme conditions result in the generation of H[•], OH[•] and HO₂[•] radicals as shown in Eqs. (27) and (28) (Pliego *et al.* 2015). The Sono-Fenton process (SFP) involves the sonolytic generation of H₂O₂ and Fe²⁺ in the presence of Fe³⁺ ions as shown in Eqs. (29)-(32) (Gligorovski *et al.* 2015).





))) – Ultrasound waves.

Synergistic effects of the sonolysis and Fenton process leads to the higher generation of OH^{\bullet} as compared to the CFP alone (Gogate 2008). In-situ production of H_2O_2 in the process saves the higher cost expenditure of reagent. However, the high energy requirements for high-frequency ultrasounds limit the application of the SFP.

3.3 Photo-Fenton process

This process involves the UV light radiations to attain the higher production of OH^{\bullet} and to regenerate the Fe^{2+} ions (Kim *et al.* 1997). The UV or visible light radiation of wavelength below 450 nm is preferably used in the process (Zepp *et al.* 1992, Mahtab and Farooqi 2020). In the process, the photoreduction of Fe^{3+} by UV irradiation causes the photochemical regeneration of Fe^{2+} , which reacts with H_2O_2 and produces OH^{\bullet} and Fe^{3+} ions as shown in Eqs. (30) and (31). The regeneration of Fe^{3+} continues the cycle and leads to higher OH^{\bullet} a production which enhanced the Fenton's oxidation performance with the results of the effective degradation of pollutants (Faust and Hoigné 1990). The process also accompanies the direct photolysis of H_2O_2 to generate the OH^{\bullet} as shown in Eq. (33). However, the presence of iron complexes in solution absorbs a large part of radiation and affects the photolysis of H_2O_2 (Safarzadeh *et al.* 1997). The role of pH is important in the photo-Fenton process (PFP) which determines the formation of different iron complexes. At a pH value of 3, the Fe^{3+} ions effectively converted into the most photo reactive ferric ion water complex i.e. $[\text{Fe}(\text{OH})]^{2+}$ species. The metal charge transfer excitation of $[\text{Fe}(\text{OH})]^{2+}$ by UV radiation regenerates the Fe^{2+} and produces OH^{\bullet} as shown in Eq. (34) (Faust and Hoigné 1990, Avetta *et al.* 2015). Acidic conditions (pH=3) also favor the conversion of carbonates and bicarbonates into carbonic acid, which comparatively exhibits low susceptibility towards OH^{\bullet} radicals (Legrini *et al.* 1993).



The addition of ligand may further enhance the regeneration of Fe^{2+} . Various ligands (L) like oxalate citrate and ethylenediaminetetraacetic etc., react with Fe^{3+} and forms Fe^{3+} stable complexes (Faust and Hoigné 1990). These complexes under the UV irradiation follow the ligand to metal charge transfer step and regenerates the Fe^{2+} ions as shown in Eq. (35). In general, the combination of photochemistry and the Fenton process is very effective than the CFP alone.

3.4. Heterogeneous Fenton process

In this process, solid Fenton catalysts are employed for the effective generation of Fe^{2+} ions. The homogeneous system of the Fenton process involves the external addition of Fe^{2+} that remains

present in the effluent of the treated wastewater and cannot be separated. Furthermore, neutralization of the Fe^{2+} in the effluent results in the formation of Fe^{3+} which eventually leads to the rise in sludge formation and needs a suitable system for its disposal. To overcome the problem of Fe^{2+} wastage, the heterogeneous Fenton (HF) process is generally preferred. In this process, the Fe^{2+} is generated from the active sites of iron oxides and other supported catalysts. The structure/pores of the catalyst keep the Fe^{2+} ions intact over its surface and immobilize them. The immobilization of Fe^{2+} reduces the ferric oxide sludge formation and extends the process feasibility over a wide range of pH. Another advantage was the reduction in loss of catalyst (Garrido-Ramírez *et al.* 2010). The process incorporates the two mechanisms for the production of highly reactive OH^{\bullet} . One way involves the direct heterogeneous Fenton reactions over the surface of the catalyst. In the other mechanism, the surface-leached iron initiates the homogeneous Fenton process outside the catalyst (He *et al.* 2016). In the direct heterogeneous Fenton process, the H_2O_2 reacts with the iron species present at the surface of solid Fenton catalysts and leads to the generation of highly reactive OH^{\bullet} . The mechanism initiated with the adsorption of organic pollutants over the surface of catalysts, these adsorbed organic pollutants then subsequently degrade through the hydroxyl radical's reactions (Pliego *et al.* 2015). The HF process also promotes the reduction of Fe^{3+} to Fe^{2+} through the electron transfer mechanisms of catalysts (Zhang *et al.* 2019, Çifçi and Süreyya 2020). Heterogeneous Fenton reactions worked well at ambient temperature and pressure conditions, which makes the reaction system more convincing (Chatzimarkou and Stalikas 2020). The most commonly used iron oxides catalyst includes maghemite, goethite, hematite, and magnetite. However, magnetite and goethite are considered to be the most widely used catalysts. Nanoparticles with their high chemical reactivity and large surface area are the other excellent heterogeneous catalysts used in the process (Pouran *et al.* 2014, ElShafei *et al.* 2017). In general, various benefits of the HF process like low iron leaching, low sludge production, the wide working range of pH, and effective regeneration of Fe^{2+} makes the process effective as compared with the CFP.

3.5 Solar photo Fenton process

The solar photo Fenton Process involves the application of solar energy to enhance the levels of hydroxyl radicals and other photoactive complexes for the higher efficiency of the Fenton process (Amor *et al.* 2015, Fernandes *et al.* 2014). The incorporation of free solar renewable energy in the Fenton process results in the high mineralization of the pollutants in very less reaction time. The solar-driven Fenton process is considered to be very efficient and achieves high mineralization (up to 90%) in a short period (about 30 minutes) (Kuo *et al.* 2010). The process is very environmentally friendly and inhibits the usage of electricity consumed in the other processes. The light irradiation considerably reduces both the reagents and electricity demands and favors higher reaction rates (Serra *et al.* 2011). Several studies conducted at the lab scale showed the impressive potential of sunlight energy as an alternative to the UV light in the photo-Fenton process (Liu *et al.* 2013, Pliego *et al.* 2014). The application of solar energy (i.e. sensitivity to wavelength less than or equal to 600 nm) in the PFP further reduces the cost involved in the process particularly for the treatment of large volumes of water (Petrovic *et al.* 2011). The solar-assisted AOP lowers the processing cost and makes the process very affordable for the commercial scale (Amor *et al.* 2015). The process with lower costs is considered to be very favorable for industrial applications (Blanco *et al.* 1999). A study conducted by Vilar *et al.* (2012) compared the treatment efficiency of the Fenton and solar-Fenton process and it was reported that the solar-assisted Fenton process (at the expense of 206 kJ L^{-1} solar energy) resulted in higher treatment efficiency with 86% mineralization and the 94% reduction of aromatic compounds of landfill leachate. The main problems encountered in the application of the solar photo Fenton process involve the amortization

costs for the design of a particular photoreactor (Pérez *et al.* 2013). The process is generally aimed at the treatment of micropollutants and industrial treatment purposes where there is a high requirement of hydroxyl radicals concentration (Malato *et al.* 2013).

3.6 Hybrid Fenton process

The Hybrid Fenton process (HFP) considered the coupling of various Fenton's approaches. It involves the combination of electrolytic, photolytic, and sonolytic Fenton processes to enhance the degradation performance of the CFP. The application of hybrid Sono-photo-Fenton leads to the higher generation of OH[•] as compared to the individual process. The hybrid Sono-photo-Fenton process involves the combined mechanism of sonolysis and UV light irradiation to enhance pollutant degradation efficiency. In the Sono-Fenton process, sonolysis of water leads to the generation of OH[•] and H[•], these radicals recombine and produce H₂O₂ which in turn reduces the OH[•] radical availability. However, the application of UV light in the hybrid Sono-Fenton process assists the decomposition of the produced H₂O₂ again into the OH[•] and thus increasing the radical's concentrations. The combined process of sonolysis and photolysis also favors the regeneration of Fe²⁺ by the reduction of intermediate complexes formed during the Fenton process (Wu *et al.* 2001). Another important hybrid technology involves the combination of photochemical and electrochemical Fenton processes. The hybrid Photo-electron Fenton process results in the higher generation of hydroxyl radicals that in turn enhances the oxidation capability of the process. The photoreduction of Fe³⁺ species via application of UV light in the EFP favors the regeneration of Fe²⁺ ions and causes the photo-decarboxylation of Fe(III)-carboxylate complexes. The photolytic hemolytic breakdown of H₂O₂ promotes the higher production of OH[•] and makes the process more efficient (Brillas *et al.* 2003, Boye *et al.* 2003). Various studies favor the applicability of hybrid Fenton processes for the treatment of landfill leachate (Altin 2008, Sruthi *et al.* 2018, Jain *et al.* 2018). In general, hybrid processes are considered to be more effective for pollutants degradation. Various other hybrid Fenton processes like Sono-electro-Fenton, Heterogeneous electro-Fenton, Heterogeneous photo-electro-Fenton, three-dimensional electro-Fenton, etc. are extensively used to enhance the degradation of the target organic contaminants. Table 1 shows several past studies for landfill leachate treatment by various Fenton-based approaches.

Table 1 Applications of the Fenton-based processes for landfill leachate treatment

Sample type	Sample characteristics	Method used	Remarks	Reference
Municipal landfill leachate	COD:1720 mg/l	Conventional Fenton process	At pH: 4, H ₂ O ₂ /Fe ²⁺ (molar ratio): 1, Fe ²⁺ : 800 mg/l and oxidation time:10 minutes, COD removal efficiency:74.7 % At optimum H ₂ O ₂ dosage: 1500 mg/l, Fe ²⁺ dosage:120 mg/l and Fe ²⁺ /H ₂ O ₂ : 0.08, COD removal: 68%	(Gau and Chang 1996)
Municipal landfill leachate	COD:2000 mg/l BOD ₅ :87 mg/l pH:8.2 BOD ₂₀ /COD: 0.10 Dark brown color	Conventional Fenton process	Decolorization efficiency: 92% with pale yellow color, BOD ₂₀ /COD improved from 0.10 to 0.58	(Kim and Huh 1997)

Table 1 Continued

Sample type	Sample characteristics	Method used	Remarks	Reference
Biologically Pretreated leachate	COD: 338 mg/l BOD ₇ : < 8 mg/l	Conventional Fenton process	At H ₂ O ₂ :10mg/l, Fe ²⁺ : 20 mg/l and H ₂ O ₂ /Fe ²⁺ : 0.8 (molar) COD removed: 72%, BOD ₇ /COD=0.3	(Welander and Henrysson 1998)
Biologically treated leachate	COD :1500 mg/l BOD ₅ /COD:0.02 pH:8.7	Conventional Fenton process	At pH=3.5 and H ₂ O ₂ /Fe ²⁺ : 14.7 (molar), COD removal efficiency: 70%	(Kang and Hwang 2000)
Pretreated leachate	COD:950 mg/l BOD ₅ /COD :0.1 NH ₃ -N:33.5 mg/l	Electro-Fenton process	At pH: 4, H ₂ O ₂ : 750 mg/L COD removal: 69%, NH ₃ -N removal :15.82 %, BOD ₅ /COD ratio increased from 0.1 to 0.29	(Lin and Chang 2000)
Biologically pretreated leachate	COD:15,700 mg/l NH ₃ -N:2,260mg/l	Fenton coagulation process	At pH:6, H ₂ O ₂ :200mg/l and Fe ²⁺ :300 mg/l, COD removal efficiency=70%	(Lau <i>et al.</i> 2001)
Biologically pretreated leachate	COD:513 mg/l BOD ₅ : 42 mg/l TOC:116 mg/l BOD ₅ /COD:0.062	Photo-Fenton process	At pH: 3-4, H ₂ O ₂ :800 mg/l and UV radiation time:30 minutes, removal efficiencies: COD: 71%, BOD ₅ : 50% and TOC: 52%, BOD ₅ /COD ratio increased from 0.062 to 0.142	(Lau <i>et al.</i> 2002)
Mature leachate	COD:10,540 mg/l BOD:2300 mg/l BOD ₅ /COD:0.2 pH:8.2	Fenton's pre-treatment process	At pH: 3, reaction time: 2 hours and H ₂ O ₂ /Fe ²⁺ : 19.7 (molar) BOD ₅ /COD ratio improved from the value of 0.2 to 0.5 and maximum COD reduction: 60%	(Lopez <i>et al.</i> 2004)
Mature leachate	COD: 5200 mg/l BOD:720 mg/l pH:8.4 BOD ₅ /COD:0.13	Photo-Fenton process	At pH: 2.8, Fe ²⁺ : 10 mg/L, H ₂ O ₂ : 2000 mg/L, COD removal: 49%, BOD ₅ /COD ratio increased from 0.13 to 0.42	(De Morais and Zamora 2005)
Landfill leachate	COD: 5000 mg/L pH: 6.4	Electro-Fenton process	At pH: 3, H ₂ O ₂ : 0.34 mol/L, Fe ²⁺ : 0.038 mol/L, COD removal efficiency:83.4%	(Zhang <i>et al.</i> 2006)
Mature municipal landfill leachate	COD: 1100-1300 mg/l pH:8.18	Conventional Fenton process	At pH: 3, [H ₂ O ₂]:0.24M and H ₂ O ₂ / Fe ²⁺ (molar ratio): 3 COD removal: 61 %	(Deng 2007)

Table 1 Continued

Sample type	Sample characteristics	Method used	Remarks	Reference
Landfill leachate	COD:2350 mg /L phosphate: 10.25 mg/ l pH: 8.36	Photo-electro-Fenton Process	pH:3, H ₂ O ₂ concentration: 3000 mg/L, current :2.5 A and treatment time:20 min, removal efficiencies: COD: 94%, phosphate: 96%, color: 97%	(Altin 2008)
Mature leachate	COD:2100 mg/l BOD ₅ /COD:0.08	Integrated Fenton-Ultrafiltration process	At pH: 3, For batch operation at reaction time: 1 hour and H ₂ O ₂ /Fe ²⁺ : 13.8 (molar), maximum COD removal:80% In continuous mode, at residence time :2 hours, H ₂ O ₂ /Fe ²⁺ : 13.8 (molar) and H ₂ O ₂ /COD:3.3 (w/w) maximum COD removal: 83%	(Primo <i>et al.</i> 2008)
Landfill leachate	COD:5700 mg/l BOD ₅ :3600 mg/l pH: 7.8 BOD ₅ /COD :0.63	Conventional Fenton process	At pH: 3.5, H ₂ O ₂ : 650 mg/L and H ₂ O ₂ / Fe ²⁺ (molar ratio): 1:19 COD removal: 66%, BOD ₅ /COD ratio increased from 0.63 to 0.88	(Kochany, and Lipczynska-Kochany 2009)
Mature municipal landfill leachate	Diluted leachate COD:93 mg/l, undiluted leachate COD:743 mg/l	Conventional Fenton process	At molar ratio H ₂ O ₂ /Fe ²⁺ =3, Fe ²⁺ dosage of 4 mmol/l, pH 3 and reaction time: 40 minutes COD removal efficiency: diluted leachate: 60.9%, undiluted leachate: 31.1%	(Cortez <i>et al.</i> 2010)
Municipal landfill leachate	COD:38200 mg/l BOD ₅ : 22000 mg/l Color (Pt-Co): 3510 pH: 7.25	Conventional Fenton process	At Fe ²⁺ :2000 mg/l and H ₂ O ₂ : 5000 mg/l, COD removal: 55.9% color removal: 88 %	(Yilmaz <i>et al.</i> 2010)
Mature landfill leachate	COD:7610 mg/l BOD ₅ :1300 mg/l BOD ₅ /COD:0.17	Photo-Fenton process	At 25 °C, pH:4, H ₂ O ₂ : 4000 mg/l, Fe (II): 30 mg/L and 60 min reaction time, COD removal: 56.3% BOD ₅ /COD ratio increased from 0.17 to 0.67	(Hu <i>et al.</i> 2011)
Stabilized landfill leachate	COD:2360 mg/l BOD ₅ :287 mg/l Iron:6.6 mg/l Color:4,200 mg/l	Conventional Fenton process	At pH: 3, H ₂ O ₂ : 0.033 mol/l Fe (II):0.011 mol/l and reaction time :145 min, COD removal: 58.3%, color removal: 79%, iron removal: 82.1%	(Mohajeri <i>et al.</i> 2011)

Table 1 Continued

Sample type	Sample characteristics	Method used	Remarks	Reference
Urban landfill leachate	COD :1573 mg/l BOD ₅ /COD:0.01 BOD ₅ : 18 mg/l	Fenton like ZVI process	At pH: 2 and H ₂ O ₂ concentration: 13.4 mg /l, COD removal: 38%, BOD ₅ /COD ratios improved from 0.01 to 0.11 At pH 8.5, H ₂ O ₂ : 3.5 g/l, Fe ²⁺ / H ₂ O ₂ : 0.25, and reaction time of 120 minutes,	(Martins <i>et al.</i> 2012)
Municipal landfill leachate	COD: 1764-2636 mg/L BOD ₅ :350–500mg /L BOD ₅ /COD: 0.157	Conventional Fenton process	COD removal: 69.8%, BOD ₅ removal: 72%, BOD ₅ /COD increased from 0.157 to 0.176	(Pieczykolan <i>et al.</i> 2013)
Sanitary landfill leachate	COD:11,280 mg/l BOD:1300 mg/l TSS:3940 mg/l pH:6.21	Integrated Coagulation-Fenton process	At pH: 7 and FeCl ₃ : 1500 mg/l, COD removal: 65%, TSS removal: 79%, color removal: 95%	(Moradi and Ghanbari, 2014)
Mature landfill leachate	COD:3800 mg/l TOC:1200 mg/l Biodegradability:0.15	Photo-Fenton process	At UV energy:110 KJ/l and H ₂ O ₂ consumption of 115 mM, COD reduction: 74 %, TOC reduction: 65% biodegradability: 0.35	(Jain <i>et al.</i> 2018)
Raw leachate	COD: 4,640±135 mgL BOD ₅ : 945±56 mg/L TOC: 1,037±69 mg/L	Sono-photo-Fenton process	At pH:3.0, H ₂ O ₂ /TOC: 2 and H ₂ O ₂ /Fe ²⁺ : 5, COD removal: 79.6%, TOC removal: 68.3%, BOD ₅ reduction: 58.2%, BOD ₅ /COD increased from 0.20 to 0.43	(Zha <i>et al.</i> 2016)
Pre-coagulated leachate membrane concentrates	COD:1120 mg/l TOC:353 mg/l	Conventional Fenton process	At pH:2, H ₂ O ₂ concentration: 1 mol /l, Fe ²⁺ :17.5 mmol/l, removal efficiencies for TOC and COD are 68.9%, 69.6%	(Xu <i>et al.</i> 2017)
Stabilized landfill leachate	BOD ₅ /COD:0.03 Catalyst: Iron-manganese oxide loaded zeolite	Heterogeneous electro-Fenton process	At pH:3, applied voltage: 4V and 25 mg/l of catalyst, COD removal: 87.5%, BOD ₅ /COD ratio increased from 0.03 to 0.52	(Sruthiet <i>et al.</i> 2018)
Stabilized landfill Leachate	COD:4852 mg/l, BOD ₅ :209 mg/l, TOC: 2894 mg/l pH: 8.56 Catalyst used: Iron Molybdophosphate (FeMoPO)	Heterogeneous Fenton process	At pH: 2, H ₂ O ₂ :9mM, and FeMoPO 1.75 g/l and reaction time :90 min, COD removal efficiency:84.9%, TOC removal efficiency:71%, BOD ₅ /COD ratio increased from 0.03 to 0.38	(Niveditha and Gandhimathi 2020)

4. COD and color reduction

The Fenton process is a widely used advanced oxidation process for COD reduction of landfill leachate in comparison with the other AOPs (Mahtab *et al.* 2021). The process is considered very advantageous in terms of its technological simplicity and cost-effectiveness (Manu and Mahamood 2011). The homogeneous treatment nature with no mass transfer limitations further favors the application of the process. The process is considered to be the most suitable choice for COD removal of leachate (Li *et al.* 2010). The results obtained from the various other studies justify the application of the Fenton process for the COD removal of leachate (Gau and Chang 1996, Welander and Henrysson 1998, Lopez *et al.* 2004, Deng 2007, Hu *et al.* 2011, Pieczykolan *et al.* 2013).

The decomposition of organic matter in the landfills (like humic and fulvic acids) may cause the percolating leachate to become yellow, brown, and black. The landfill leachate is usually dark in color (Aziz *et al.* 2007). The Fenton process is considered an effective choice for the reduction of the color of landfill leachate. Yilmaz *et al.* (2010) conducted the study for leachate treatment by the Fenton process and reported that at 5000 mg/l of H_2O_2 and 2000 mg/l of Fe^{2+} , the reduction of color was 88%. Mohajeri *et al.* (2010) also evaluated the effectiveness of Fenton's technique for the treatment of semi-aerobic landfill leachate and reported the color reduction of 78.3% under the optimum operating conditions of pH 3, $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ molar ratio of 3, and reaction time of 120 minutes. Another study performed by Kim and Huh (1997) on the Fenton process for landfill leachate showed the effectiveness of the process in terms of color reduction. This study reported a decolorization efficiency of 92% with a noticeable change in the color of leachate from dark brown to pale yellow.

5. Increase in biodegradability

Landfill leachate is usually characterized by the presence of high recalcitrant or non-biodegradable organic compounds. The biological processes are not yielding efficient results for the removal of these compounds. The Fenton process is considered to be an effective alternative to oxidize these compounds and increase the biodegradability of the leachate for the associated biological treatment processes to meet the standard discharge limits. A study performed by Kim and Huh (1997) on the Fenton's treatment for landfill leachate concluded that at the optimum dosage of 1500 mg/l of H_2O_2 and 120 mg/l of Fe^{2+} , the biodegradability of leachate increased from the value 0.10 to 0.58. Kochany and Lipczynska (2009) in their study reported the enhanced biodegradability of landfill leachate from the value of 0.63 to 0.88 by the Fenton process, at the optimized conditions of pH 3.5, 650 mg/l of H_2O_2 . Another study conducted by Lopez *et al.* (2004) reported that at pH 3 with an $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ molar ratio of 19.7 and reaction time of 2 hours in Fenton's treatment of municipal landfill leachate, the BOD_5/COD ratio increased from the value of 0.2 to 0.5 and hence increased the biodegradability of the leachate.

6. Removal of Humic Acids from leachate

The landfill leachates are generally characterized by the presence of a high concentration of humic and fulvic acids. These acids are considered to be very recalcitrant and do not degrade easily (Wang *et al.* 2014). It is reported that the concentration of fulvic acids decreases over time,

however, the concentration of humic acids goes on increasing with time (Artiola-Fortuny *et al.* 1982). Humic acids are found to be in very high concentration in both the mature and old landfill leachates (Wiszniewski *et al.* 2004). The humic acid acids tend to bind with the heavy metals and organic contaminants like herbicides, insecticides, and pesticides present in the landfill leachate and takes away these pollutants from landfills to the surrounding environment (Calace *et al.* 2001). Hence the removal of humic acids is considered to be necessary for the treatment of landfill leachate. The Fenton process is considered to be one of the very effective technology for the removal of humic acid concentration from landfill leachate. A study conducted by Wu *et al.* (2011) investigated the application of the Fenton process for the removal of humic acids and it has been found that the low initial pH values and adequate dosages of absolute and relative Fenton's reagent dosages has resulted in the high removal of humic acids. Another study conducted by Sarmiento *et al.* (2018) suggested that CFP is a very effective technique for the degradation of humic acids. It has been reported that at the pH value of 5, Fe^{2+} of 0.54 mmol/l, H_2O_2 of 6.17 mol/l, and a reaction time of 120 minutes, the effective removal of humic acids was achieved.

7. Combined processes

The application of CFP with other technologies like UV radiation, sonolysis, and electrolysis, etc, results in the higher generation of hydroxyl radicals that ultimately lead to the higher degradation of pollutants. The combined processes enhanced the degradation rate of the target organic contaminants. Other advantages may include the low sludge production, in-situ generation of H_2O_2 , and regeneration of ferrous ions. A study conducted by Lau *et al.* (2002) showed the effectiveness of the PFP for the treatment of landfill leachate with reported results of 71%, 50%, and 52% of reductions in the COD, BOD, and TOC concentrations, respectively. Moradi and Ghanbari (2014) in their study evaluated the efficiency of integrated coagulation Fenton process for the landfill leachate treatment. The process came out to be very effective in terms of the COD, TSS, and color reduction of leachate. At the optimized treatment conditions, the removal of 65% of COD, 79% of TSS, and 95% of color was achieved. Niveditha and Gandhimathi (2020) studied the effectiveness of the HF process for the treatment of landfill leachate. The process showed the removal of 84.9% COD and 71% of TOC. The treatment was further observed with the enhanced biodegradability of the leachate. The hybrid process focused on the coupling of electrolytic, photolytic, and sonolytic processes to accomplish the faster degradation rate as compared to the individual photo, electro, or Sono-Fenton systems. Altin (2008) evaluated the effectiveness of the hybrid Fenton process for the treatment of landfill leachate. In the combined photo-electron-Fenton process used in the study, the obtained results were a reduction of 94% of COD, 94% of phosphates, and 97% of color.

8. Pre-treatment and post-treatment options

Fenton's approaches are extensively used as pre/post-treatment of landfill leachate. The pre-treatment option enhances the biodegradability of the leachate and makes it suitable for subsequent biological treatments. The biological processes are effective for the treatment of wastewater with a comparatively high BOD_5/COD ratio. On the other hand, the post-treatment option results in further COD removal and increased the biodegradability of effluents to meet the standard

discharge limits. A study conducted by Lau *et al.* (2002) evaluated the post-treatment efficiency of the photo-assisted Fenton system followed by the up-flow anaerobic sludge blanket (UASB) treatment of landfill leachate. As a post-treatment method, it reduced the COD of leachate from an average value of 1910 mg/l to 513 mg/l and also enhanced the biodegradability of the leachate from the value of 0.062 to 0.142. Lopez *et al.* (2004) conducted the study to check the effectiveness of the Fenton process for the municipal landfill leachate. The Fenton's treatment enhanced the BOD₅/COD ratio of leachate from the value of 0.2 to 0.5 to meet the required conditions for the subsequent biological treatments. The pre-treatment option was also favored with a 60% of COD reduction. Wang *et al.* (2000) studied the two-stage treatment process of landfill leachate by employing the UASB and Fenton process. In the second stage, the Fenton coagulation process removed about 70% of COD present in the UASB pretreated leachate, lowered the value of 1500 mg/l to 447 mg/l. The low COD value of the effluent satisfied the discharge standards. A study conducted by Poblete and Pérez (2020) considered the adsorption process involving the use of sawdust for the treatment of landfill leachate followed by the PFP. The sawdust adsorption treatment of landfill leachate resulted in an 18.3% reduction of humic acids, 33.7% reduction of COD, and a 19.5% reduction of color. It has been reported that the adsorption process caused the reduction of ammonium and heavy metals present in the leachate. Another study conducted by Jaafarzadeh *et al.* (2016) investigated the chemical precipitation process for pretreatment of mature landfill leachate followed by the Fenton's oxidation process. The results of this study show that chemical struvite precipitation followed by the Fenton oxidation resulted in 87% of NH₄ removal, and about 95% removal of both the COD and BOD. Hence the study suggests that the struvite chemical precipitation is considered to be a very efficient, reliable, and feasible pretreatment alternative for Fenton's oxidation treatment of mature landfill leachate. In another study, Kanaani *et al.* (2019) has adopted the coagulation process as a pretreatment scheme for the solid waste compost leachate treatment before the Fenton process and it has been observed that at the coagulation stage, about 45% of COD reduction has been achieved. Furthermore, the pretreatment followed by the Fenton process resulted in a total reduction of 98.2% of BOD and 84.4% of COD. Suresh *et al.* (2016) studied the Membrane bioreactor (MBR) technology for the pretreatment of landfill leachate before the final treatment by the EFP. Results showed that after the MBR pretreatment process, the levels of COD, BOD, ammonia nitrogen have been reduced by 75, 79, and 74% respectively. The application of the EFP after the MBR treatment further resulted in the reduction of 77% of COD, 74% of BOD, and 88% of ammonium nitrogen concentrations. Hence it was concluded the MBR process is quite an effective option for the pretreatment of landfill leachate followed by the EFP.

9. Conclusions

After going through a wide literature survey, we can say that the Fenton-based approaches are one of the most attractive and effective choices among AOPs for removal of a large spectrum of refractory compounds present in the landfill leachate, COD, BOD, color, TOC, TSS and improving biodegradability, etc. The easily available and non-toxic nature of Fenton's reagents (i.e. H₂O₂ and Fe²⁺) further favors the extensive use of the CFP. The process can be made more effective by associating it with other technologies like sonolysis, photolysis, and electrolysis, etc. The combined process relieves the problem of sludge generations assists the regeneration of Fe²⁺ and leads to the higher generation of hydroxyl radicals, ultimately improved the overall treatment

efficacies. However, each advanced approach is having some limitations, no single process is sufficient to serve the overall purpose. Thus, we have to suitably select the Fenton-based approaches either single or in combinations according to the characteristics of the sample, available resources, and required discharge standards. The choices should be cost-effective, environmentally friendly as a sustainable solution to the problem.

About 75% of COD can be removed by the CFP while the hybrid Fenton processes can go up to about 95 % of COD removal of the landfill leachate. The hybrid Fenton's treatment remarkably removed the color up to 97%. Furthermore, the process enhanced the biodegradability of leachate as well, which favors further biological treatments. Various factors like pH, temperature, concentrations of H_2O_2 , the dosage of Fe^{2+} , optimum reagents ratio (H_2O_2/Fe^{2+}), nature of reaction matrix, and reaction time influence the oxidation mechanism of the process. pH and Fenton's reagents ratio are the crucial factor that determines the overall efficiency of the process in many ways. An excessive dosage of H_2O_2 and Fe^{2+} leads to the scavenging of hydroxyl radicals and hinders the process efficiency. Thus, it is mandatory to optimize all the operational parameters very carefully for efficient treatment, which also reduces the restrictions associated with the processes. Further studies are required to overcome the limitations involved in each Fenton-based approach as discussed above, to make the process more user-friendly, efficient, economical, and environmentally friendly. As a sustainable solution for high strength wastewater treatment that can be utilized also for large-scale applications. Besides, for enhanced organics removal, integrated processes would be more suitable hence more research should focus on it.

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