

Municipal solid waste leachate treatment using ultrasonication microwave & Ozonation combined Fenton process

V. Gowri Maheswari, M. Angelina Thanga Ajisha & Jaslin J. Christopher

To cite this article: V. Gowri Maheswari, M. Angelina Thanga Ajisha & Jaslin J. Christopher (2021): Municipal solid waste leachate treatment using ultrasonication microwave & Ozonation combined Fenton process, Australian Journal of Civil Engineering, DOI: [10.1080/14488353.2021.1941596](https://doi.org/10.1080/14488353.2021.1941596)

To link to this article: <https://doi.org/10.1080/14488353.2021.1941596>



Published online: 06 Aug 2021.



Submit your article to this journal [↗](#)



Article views: 15



View related articles [↗](#)



View Crossmark data [↗](#)

Municipal solid waste leachate treatment using ultrasonication microwave & Ozonation combined Fenton process

V. Gowri Maheswari^a, M. Angelina Thanga Ajisha^b and Jaslin J. Christopher^c

^aCivil Engineering Department, Francis Xavier Engineering College, Tirunelveli, India; ^bCivil Engineering Department, SCAD College of Engineering and Technology, Tirunelveli, India; ^cChemistry Department, Manonmanium Sundaranar University Constituent College, Kanyakumari, India

ABSTRACT

This work focus on the effect of Microwave power, Ozonation and Fenton reagent in microwave coupling Ozonation and Fenton oxidation process for the disintegration of COD of leachate. The experimental parameters like pH, COD, temperature, Oxidant dosage, microwave power and reaction time were varied. The results showed that 70% of COD removal efficiency was achieved at 450 W within 10 minutes interval in microwave process, 57.4% of COD removal efficiency was achieved at 60 minutes, ozone dosage 159 mg/L, pH 11. In Fenton Process, 52.91% COD removal efficiency at 60 minutes with H₂O₂ 66.47 mμ/L and Fe²⁺ 80 mμ/L. Individual process showed an adverse reaction time consumption. On comparison of three processes, microwave achieved higher COD removal with shorter reaction time. In O₃+ Fe process 83% COD removal was achieved at 75 minutes of reaction time. The experiment revealed the combined process of M+ O₃+ Fe induces to the disintegration of COD in Leachate with a shorter reaction period.

ARTICLE HISTORY

Received 9 May 2020
Accepted 8 June 2021

KEYWORDS

Leachate; ultrasonic; microwave; Fenton; Ozonation

1. Introduction

The landfilling of municipal solid waste (MSW) is a major problem in developed and developing countries (Kjeldsen et al. 2002). MSW can be classified into residential, commercial, food, bulk, wood, dead animal, hazardous waste and industrial un-hazardous waste. It consists of both bio-degradable and non-biodegradable materials. Disposal of MSW releases a liquid known as leachate.

Leachate is generated from various chemical reactions which occur due to the degradation of organic matter as well as percolation of precipitated waste entering into the landfill. Improper design, management and operation of MSW cause serious threats to the environment such as soil contamination, air pollution, surface and ground water contamination. It is reported that 100,000 Million Tonne (MT) of waste is generated in India (Rocha et al. 2011). Waste generation in Tirunelveli city was estimated to be 161.44 MT. Hence, there is a need for the treatment of leachate before discharging into the environment. There are various treatment techniques adopted for landfill leachate treatment such as aerobic and anaerobic biodegradation, micro algal treatment, ultrasonication, leachate transfer, coagulation & nano-filtration, catalytic oxidation, electrochemical oxidation, electro dialysis, Fenton

process and Ozonation. It was reported that AOP has several advantages such as easy installations, low cost, low energy requirement, shorter reaction time, elimination of colour and increase in biodegradation ability of recalcitrant substances (Deng and Englehardt 2006). Most of the process in AOP commence on the production of free-radicals especially hydroxyl species. This free radical production can be achieved either by a single process (Ozonation) or by the combination of various strong Oxidants such as ferrous ion and Hydrogen peroxide.

The photo Fenton process can be enhanced by employing UV radiation that reduces the dissolution of Fe³⁺ ion into Fe²⁺ complexes and generation of Hydroxyl radicals to photolysis process (Boyle and Ham 1974). It was reported that Ozone treatment can be effectively adapted for the treatment of complex leachate generated from MSW (Ho, Boyle, and Ham 1974).

Ozone treatment is highly effective at elevated pH which leads to the generation of Hydroxyl radical is due to the serious complex chemical reactions between OH⁻ ion and Hydrogen peroxide. Ozone and Hydrogen peroxide produces 1 mol of Hydrogen peroxide for 1 mol of Ozone compared to 1.5 mol of Ozone in the case of pure Ozone.

2. Materials and methods

2.1. Characterisation of MSW leachate

Ramayanpaati landfill site is located in Tirunelveli city, Tamil Nadu, India. The landfill has an operational area of 20 acres with an annual average of rainfall of 680 millimetres (27 in.). The leachate samples were collected with poly-ethylene bottles and the collected sample was immediately transported to the laboratory and refrigerated at 4°C as per the standard methods. The initial characteristics of leachate were pH = 8.57, Temperature = 33°C, Total solids = 16,350 mg L⁻¹, Total suspended solids = 2580 mg L⁻¹, Total dissolved solids = 13196 mg L⁻¹, Alkalinity = 10,500 mg L⁻¹, COD = 39333.33 mg L⁻¹, BOD = 284.4 mg L⁻¹, Nitrogen = 375.2 mg L⁻¹ and Turbidity = 2778.7 NTU.

2.2. Materials

The Photo-Fenton process was carried out by using Ferrous Sulphate (FeSO₄.7H₂O), Hydrogen peroxide (H₂O₂ - 50%), Hydrochloric Acid (HCl - 33%) and Sodium Hydroxide (NaOH-30%) (for pH adjustment). All the chemicals were purchased from Merck, Mumbai.

2.3. Experimental methods

The Microwave used for the study is the IFB 30SC2 model with an output power of 900 W. The reactor volume is 1000 mL. It has a frequency of 2450 MHz. 1000 mL samples were used for conducting the experiments. The power of the microwave varied from 90 to 450 W and the sample was collected at 5, 10, 15, 20, 25 and 30 min.

2.4. Experimental procedure

2.4.1. Microwave

The Microwave process conducted on an IFB made with a total volume of 30 L. The microwave apparatus produced 900 W at a frequency of 2450 MHz, The apparatus was set to rise the temperature of the sample from 32°C to 85°C within 15 minutes during the experimental condition and kept constant with various Microwave powers.

2.4.2. Fenton process

The Laboratory Photo-Fenton experiment was conducted in a batch reactor at room temperature and at atmospheric pressure in a laboratory glass reactor with a working volume of 1.5 L. The pH of the sample was adjusted with the help of sulphuric acid and sodium hydroxide. The pre-determination of Fe²⁺ dosage was achieved by the addition of required amount of ferrous sulphate (35%). Hydrogen peroxide solution with a calculated volume was added. After a fixed detention

period of 60 minutes the Leachate was allowed to settle for 1 hour and the sample was analysed. All the experiments were triplicate and the results were averaged.

2.4.3. Ozonation

The Ozonation experiment was conducted in a glass reactor with a volume of 1 L feed with Ozone with a glass centre diffuser. The Ozone was generated using a Faraday Ozone generator with dry pure Oxygen and the gas flow rate was 0.5 LPM. The Ozone concentration was measured using a UV gas analyser. The sample was immediately withdrawn and allowed to settle for 30 minutes for analysis. The effect of pH values of Ozonation experiments were carried out using sodium Hydroxide to achieve the required values of 8–11. All the experiments were conducted at room temperature and the experiments were triplicate and the results were averaged.

3. Result and discussions

3.1. Effect of micro wave power setting

Treatment efficiency of landfill leachate with micro wave variation is depends upon the dielectric property of leachate and power applied in terms of micro wave variation. Yo-chich et al. reported that leachate has a high dielectric constant due to water content and it can be heated up well. The following relation relates the micro wave energy absorbed by the leachate to the power and irradiation time.

$$P_{abs}X_t = B * C_p * M * X_t \quad (1)$$

Where,

$P_{abs} X_t$ = Micro wave energy absorbed by the leachate.

P_{abs} = Micro Wave power.

B = Unit conversion coefficient.

C_p = Heat capacity.

M = mols of the fluid.

It was observed that energy applied in the form of microwave is proportional to the average temperature rise to the desired level (85%). It is concluded that a higher increase in the microwave power will increase the temperature in a shorter period of time. Microwave treatment is more effective as compared to that of conventional heating due to the following reasons:

Higher heating rate.

Reduction of activation energy.

Reduction of reaction rate and size.

Energy efficiency.

Microwave treatment of leachate possesses volumetric heat property and superior process control. The energy efficiency of COD, TOC reduction in the leachate can be expressed as

$$\beta = \text{COD}[\text{or TOC}] * V * r^2 / (P * t) \quad (2)$$

β – Energy efficiency associated with COD or TOC in leachate mg (COD or TOC)/ (KW-h)

COD /TOC – Initial COD /TOC value

V – Volume of leachate in litres.

r^2 – COD removal rate

P – Microwave irradiation power

T – Irradiation time

In this study, the leachate was raised to 85°C from the initial temperature of 32°C with various power settings such as 90, 180, 270, 360, 450 W and time. Figure 1 shows the relationship between COD removal and reaction time with various powers applied for leachate treatment. When the power was increased from 90 to 450 W, a 58% increase in the COD removal efficiency was observed within 10 minutes. When the irradiation time was increased to 5 minutes there was a reduction of 24% in the COD. The increase in irradiation time of more than 15 minutes for various power settings showed no significant improvement in COD reduction. It is also observed that based on the Equation (3). $P_{\text{abs}} * t$ is proportional to the mass of the fluid.

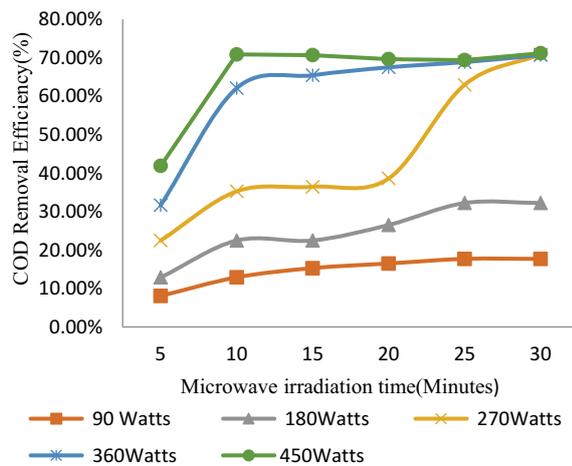


Figure 1. Effect of microwave irradiation time on COD removal efficiency.

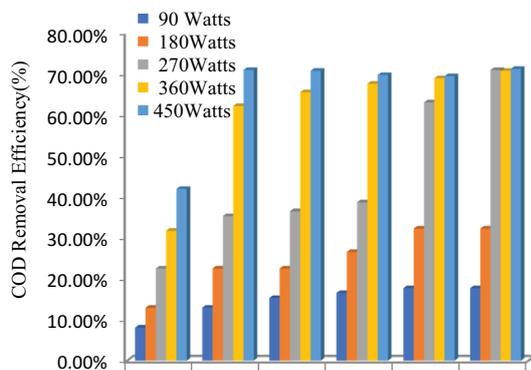


Figure 2. Effect of microwave power on COD removal efficiency.

$$\frac{M \times \Delta t}{P_{\text{abs}}} = t \quad (3)$$

Based on Equation (1), it was observed that high power with short irradiation time is required to elevate the temperature to 85°C. The same result was observed from in this study. This COD reduction in leachate is due to the difference in dielectric loss tangent ($\tan \Delta$).

At a given temperature the ability of the material to store electric energy must be the same in order to achieve conversion of microwave energy into the heat the dielectric loss factor must be increased. This can be achieved by increasing microwave power. It was concluded from the experiment that microwave energy applied to the leachate and COD removal can be thermal and athermal. The thermal effects contribute to high rise in the temperature of the leachate and molecular vibration the athermal process due to the production of free radicals and oxidation of substance, it implies that higher microwave power provides more energy input and faster molecular vibration which increases the reaction rate which can be observed in Figure 1.

At lower microwave energy input the removal efficiency increases from 8% to 17% at 30 minutes intervals. However when the power is increased to 270 W at 30 minutes the removal efficiency increases to 70% for the same removal efficiency if the power increased to 450 W. It achieves 70% efficiency within 10 minutes intervals. After 10 minutes of microwave irradiation TOC reduction was observed as 70.63%. However, the efficiency of COD reduction was very low at power reducing from 360 to 90 W (reference). In order to understand the effect of microwave power on COD reduction in leachate the method adopted by Yu-Chieh-Choa, the following methods of combination was adopted.

- Raw leachate
- Raw leachate + Microwave

3.2. Ozonation process

3.2.1. Effect of time on Ozonation efficiency

Time was an influential parameter in the reduction of COD of the landfill leachate in Ozonisation. To understand its effects, the leachate samples were ozonated for 30 minutes and 60 minutes and the results are shown in Figure 3, it was found that the increase in the Ozonation time increases the removal efficiency after 50 minutes and increase in the concentration of ozone increases removal efficiency up to 46% after 50 minutes of Ozonation. This increase in COD reduction was observed due to the oxidation of aromatic and carbon double bonds.

From the Figure 4 it was observed that the lower concentration of ozone supply to the leachate

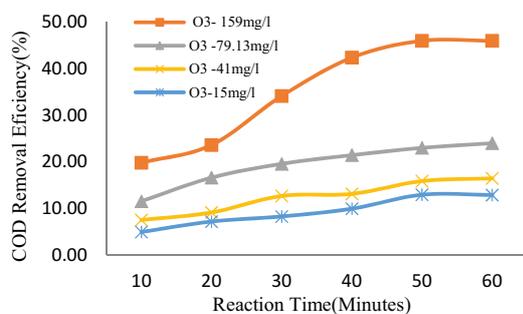


Figure 3. Effect of ozone reaction time on COD removal efficiency.

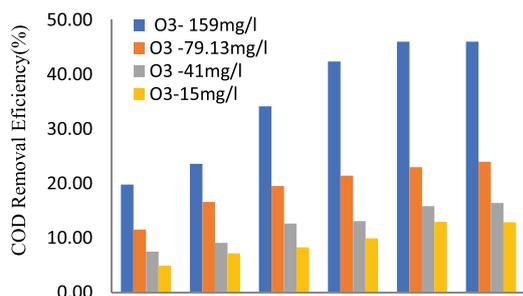


Figure 4. Effect of ozone consumption on COD removal efficiency.

produces only 12.85% removal. This phenomenon may be due to the presence of high recalcitrant substance other, organic and inorganic pollutants. Furthermore, ozonisation at lower concentrations produced intermediate derivatives which also cause a reduction in the overall COD value in the leachate after 60 minutes. Further, the increase in the concentration for a fold of 10 times (159 mg/L) increases the transfer rate of ozone and easily oxidises the compounds present in the liquid. Hence, high removal of COD was observed.

3.3. Effect of pH

The correlation between COD removal and the initial pH of the leachate is shown in the Figure 5. From the results it was observed that the highest reduction of COD was achieved at pH 11.

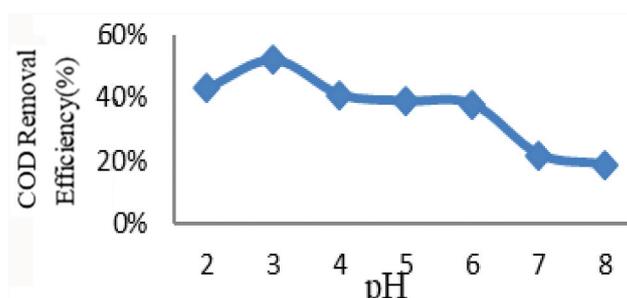


Figure 5. Effect of pH on COD removal efficiency.

Table 1 shows the amount of ozone transfer to the leachate solution at various pH with respect to time the phenomena of improvement in the efficiency of COD reduction at alkaline pHs was due to difference in the amount of ozone transferred to the leachate solution and subsequent ozone decomposition rate and the highest amount of ozone consumption at pH 11 leads to the formation of increased hydroxyl radicals due to the reaction ozone at hydrogen ion present in the leachate. Hydroxyl radicals are non-selective towards unsaturated, complex large molecules hence highest COD reduction was achieved at pH-11 after 60 minutes of treatment time. The reduction of COD at neutral pH (8.1) was observed compared to pH-11 for ozonisation the process mainly depends on the presence of molecular ozone and the presence of hydroxyl radicals is most essential. However, in the pH-8 the strong oxidant ozone is selectively reacting with leachate at lower reaction rate than hydroxyl radicals. Moreover, at neutral pH equilibrium reaction of organic pollutants turns towards non-ionised forms compared to ionised and dissociated forms. This makes the ozone less reactive selectively for those compounds.

3.4. Fenton process

3.4.1. Effect of pH

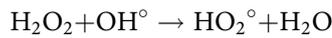
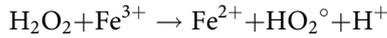
Fenton process will effectively operate at a narrow pH range the pH of the leachate affects the activity of oxidant and leachate ions species and hydrogen peroxide. Zhang et al reported that at pH ranging from 2 to 4, which effectively yields hydroxyl radical production from hydrogen peroxide which reacts with organic-metallic complexes. Inorganic carbons are scavengers of hydroxyl radicals that can be effectively removed by producing pH in the acidic range. Figure 5 shows the influence of pH on COD removal using Fenton process from the plot it was observed that the optimum pH from the present study is 3 with a removal efficiency of 52%. At optimum pH the soluble ion species is hydrated ferrous ion $[(Fe(H_2O)_4)^{3+}]$.

The following stoichiometric reaction shows the mechanism of reduction of hydrogen peroxide and

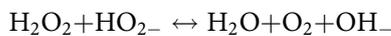
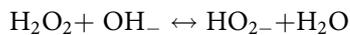
Table 1. Effect of pH for COD removal in Ozonation process.

Time	COD Removal Efficiency pH-8	COD Removal Efficiency pH-9	COD Removal Efficiency pH-10	COD Removal Efficiency pH-11
10	20.22	19.5	21.95	21.68
20	29.4	30.5	31.26	30.9
30	35.8	38.91	36.78	38.11
40	44.4	46.03	48.29	47.62
50	51.48	54.61	55	57.67
60	51.48	54.5	54.8	57.4

ferrous ion production and hydroxyl radical production.



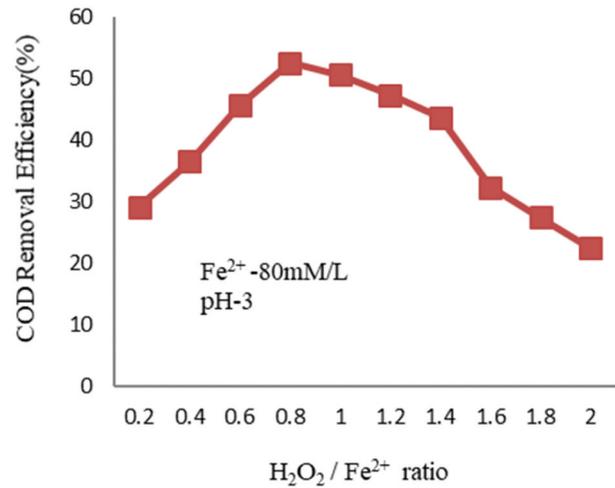
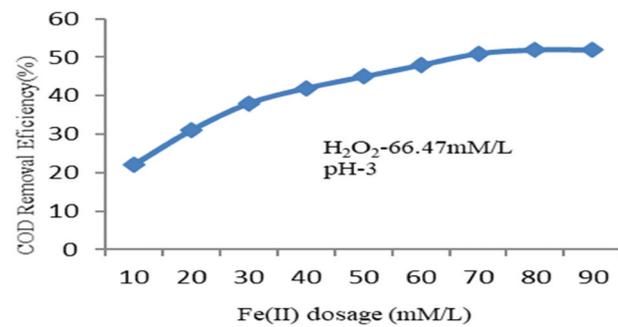
Hydrogen peroxide decomposes at optimum pH and reacts with soluble ion concentration by improving oxidation. Based on the equation when the pH increases, the concentration of hydrogen peroxide residual also increases. This mechanism reduces the production of hydroxyl radicals. The reaction of soluble ion reacts with hydrogen peroxide was decreased thus increases the residual concentration of hydrogen peroxide. This increase in the hydrogen peroxide concentration produces oxygen due to less amount of ion concentration at particular pH range (alkaline) (Lin, Zhang, and Hou 2014)



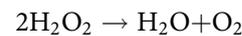
3.4.2. Effect of hydrogen peroxide concentration

Hydrogen peroxide plays a major role in the treatment of the Fenton process. The concentration of hydrogen peroxide depends upon the stoichiometric ratio of hydrogen peroxide and the initial COD of the leachate. In this work different dosage of H_2O_2 / Fe^{2+} ratio ranging from 0.2 to 2.4 was investigated. To understand the optimum ratio for treatment, the pH and Fe^{2+} dosage values were kept constant and the experiment was conducted for different H_2O_2 concentrations. $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ molar ratio of 0.8 gives the maximum COD removal efficiency. Based on the graph it was found that increase in the H_2O_2 concentration increases the COD removal efficiency to 56.56%. The maximum COD removal efficiency was obtained at an H_2O_2 dosage of 66.47 mM/L.

Based on Figure 6, it was observed that the increase in the COD removal was influenced by the addition of H_2O_2 this may be due to the production of hydroxyl radicals. However, increase in the hydrogen peroxide dosage of more than 66.47 mM/L produces different levels of the reaction rate, further increase in the H_2O_2 dosage reduces the COD removal efficiency. This may


Figure 6. Effect of $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ ratio on COD removal efficiency.

Figure 7. Effect of $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ ratio on COD removal efficiency.

be due to the decomposition of H_2O_2 into oxygen and water.



The excess amount of H_2O_2 beyond the optimum level combines with the OH° radical and organic pollutant. This process makes H_2O_2 as OH° radical scavenging capacity. Excess amount of H_2O_2 causes ion sludge flotation and auto decomposition of H_2O_2 due to oxygen off-gas.

The role of Hydrogen peroxide and ferrous ion in the treatment of landfill leachate by Fenton helps to determine optimal dosage. Hydrogen peroxide dosage is highly critical and it affects the theoretical maximum mass of OH° generated. Hence, if the initial COD is high the dosage of hydrogen peroxide will be more. To evaluate the efficiency of hydrogen peroxide usage in the leachate treatment using the Fenton process

$$\eta = 2.12 \text{ COD}_{\text{oxi}} / [\text{H}_2\text{O}_2]$$

Kim et al. (1997) reported that for untreated leachate with low H_2O_2 added concentration related to COD the η value may be above 100 and he also suggest that the organic substances present in the leachate was primarily oxidised by OH° radical formed by H_2O_2 . However, the decrease in the η value below 100% by increasing H_2O_2 dosage indicates that the substances

present in the leachate are organic matter with refractory elements (Kang and Hwang 2000).

3.4.3. Effect of ferrous sulphate concentration

The Fenton process primarily depends on the operational parameters such as $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ and organic matter/ Fe^{2+} ratio. In order to investigate the optimum ratio of $\text{H}_2\text{O}_2/\text{Fe}^{2+}$, different concentrations of $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ molar ratios were tested by keeping H_2O_2 and pH as constant. The optimum dosage of $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ was observed to be 0.8 and it further increases the optimum dosage which indicated no significant improvement in the COD removal efficiency. This may be due to the scavenging effect of H_2O_2 on OH° radical and the formation of hydroperoxyl radical [HO_2°], which is less attractive and reactive with the recalcitrant substance available in the leachate and not allowing the contribution to oxidation to produce the pathway yielding carbonate, chlorides and sulphate (Badawy et al. 2013).

In the Fenton process, the amount of Fe^{2+} salt concentration is highly important to maximise the net production of OH° radical (Kang and Hwang 2000). From our research it was observed that a lower ratio of $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ was observed and selected as optimum value because lower Fe^{2+} will contribute to the reduction in the treatment cost and lower sludge production. The optimum dosage of $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ was observed with a removal efficiency of 52% at the optimum pH of 3. Further increase in the dosage of $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ cannot modify any changes in the residual COD degradation. This fact may be due to the end by-product of Fenton reactions or mainly made up of short chain organic acids which are difficult to oxidise further.

At optimum dosage the maximum amount of COD removal was observed. The result may be due to the production of OH° radical and formation of colloidal particles due to polymerisation or condensation of organic matter which shifts the sludge distribution towards larger molecule and improvement of ferric ion complexity towards oxidised organic matter resulting in easier precipitation (Lopez et al. 2004).

3.4.4. Effect of contact time

The experiments were conducted to understand whether the reaction time is greater than 60 minutes in order to find out the beneficial COD reduction and reaction design which steps to extended the leachate oxidation. Figure 8 shows that there is no further oxidation taking place in the reactor and a further increase in the reaction time from 60 minutes to 75 minutes there is no significant COD removal efficiency. When the leachate is oxidised by the Fenton reagent the primary degradation

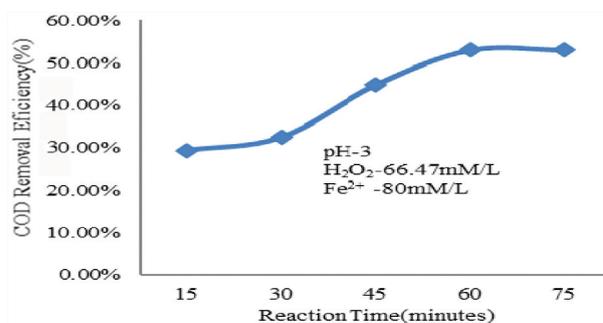


Figure 8. Effect of reaction time on COD removal efficiency.

taking place which changes the structure of the compound and makes them bio-degradable. TOC in this work is measured since TOC is independent of the oxidation state of the leachate organic matter and it doesn't measure any organic bond elements to contribute oxygen demand in the Fenton process. The leachate organic matter degrades to carbon dioxide and water and hence TOC may be decreased. However, any structural change of organic matter degradation of toxicity of organic matter occurs, TOC remains constant. The relationship between oxidation state in leachate relating with COD and TOC is expressed as,

$$\text{Oxidation state} = \frac{4 \times \{\text{TOC} - \text{COD}\}}{\text{TOC}} \quad (4)$$

Where,

COD is expressed in terms of mols of oxygen per litre

TOC is expressed in mols carbon per litre

It was found that during the first 15 minutes the COD removal efficiency was found to be 30% and further a increase in the reaction time increases the removal efficiency twice the amount observed at 30 minutes for COD. (Zhang et al. 2015)

Figure 8 shows the change in the oxidation state of organic carbon in the landfill leachate during the initial 15 minutes of time. The oxidation state was rapidly increasing and however after 60 minutes, the levels of and the mean oxidation state of organic matter in the landfill leachate was found to be 52.91%. It was concluded that the organic material present in the leachate reacts with Fenton reagent and converted into highly oxidised by-product. Further, the ultimate conversion of organic carbon compound into inorganic carbon was not significant and further increase in the hydroxyl radical concentration increases the carbon compound into partially oxidisable compound than carbon dioxide. This was concluded from the measured values of observed of residual ferrous ion and hydrogen peroxide. Hence, the reaction time for the Fenton treatment the reaction time was observed to be 60 minutes (Deva et al. 2020; Krishnan et al. 2015b, 2015a).

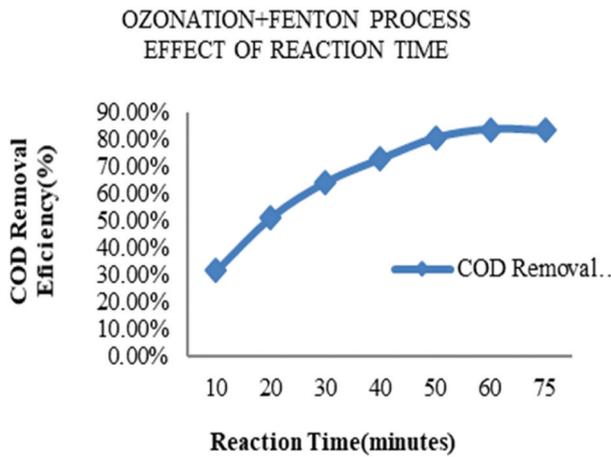


Figure 9. Effect of reaction time in Ozonation-Fenton process.

3.5. Combined scheme of Ozonation-Fenton for leachate treatment

Figure 9 shows the result of combined ozone and Fenton process for COD reduction in landfill leachate. The result observed from our study indicates that there is improvement in the performance in COD reduction of 30% than Ozonation and Fenton process alone. The combined treatment improves the biodegradability of landfill leachate. During the treatment of ozonisation, there is a slight increase in the pH from the initial pH value of landfill leachate. This increase in the pH may be due to the formation and stripping of carbon dioxide and volatile fatty acids (Krishnan et al. 2010).

When the ozone pre-treatment was adopted without adjusting the natural pH the removal efficiency was 51.48%. On the other hand, when the Fenton pre-treatment was adopted the removal efficiency was observed to be 19%. This result shows that the landfill leachate can be effectively pre-treated with ozone without adjusting the pH compared to Fenton process. However, when the pH was reduced to the optimum value of pH-3 using Fenton pre-treatment the removal efficiency was 1.43% increase than the ozone pre-treatment. When the ozone pre-treated leachate was subjected to Fenton treatment at optimum pH the removal efficiency was increased to 78.48%, 26.57% and 28% higher than individual Fenton and Ozonation process, which is shown in Figure 10.

During the ozonisation process, the biodegradability of landfill leachate was observed and found that Ozonation alone converts the organic compound into a bio-resistive compound due to primary mineralisation (Goi, Veressinina, and Trapido 2009). When the combined treatment was adopted at pH 8 improvement of biodegradability was observed (BOD/ COD- O3 @ pH 8 Fe at pH 8). When the Ozonation treatment was adopted at an optimum pH of 11

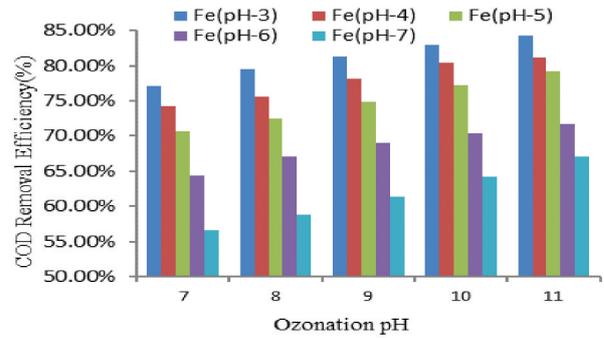


Figure 10. Effect of different pH on COD removal efficiency.

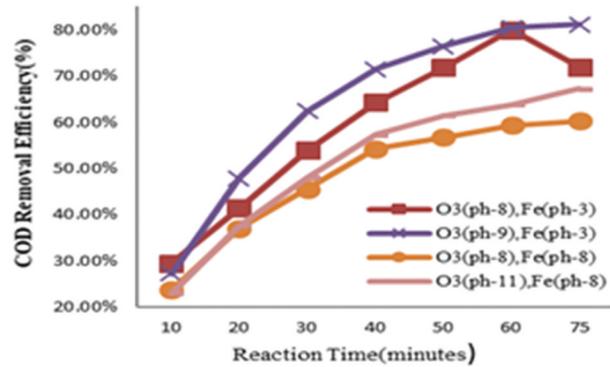
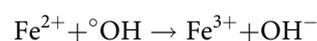
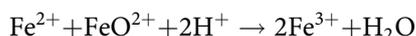


Figure 11. Effect of pH on COD removal efficiency in Ozonation-Fenton process.

followed by Fenton process at pH 3 the removal efficiency was observed to be 84.25% which is 32.25% and 26.57% greater than Fenton and Ozonation alone in the respective optimum pH of 11, 3. Moreover there is an increase in the efficiency of 32.47% and 28.58% than the treatment adopted at natural pH of the leachate with Ozonation alone and combined pre-treatment of Ozonation and Fenton. This result shows the combined treatment process Ozonation-Fenton was more efficient compared to Ozonation after Fenton process reported by Goi, Veressinina, and Trapido (2009) and several studies reported that high bio-degradation and better removal efficiency of landfill leachate using combined Ozonation-Fenton process by Anna Goi et al., E. De Torres-Socias et al., S.S. Abu Amr et al. It was observed that slight decrease in the pH was observed during the treatment of Ozonation. pH decrease was observed during the experimental conditions and reported by many researches, such as Nawrochi et al. (2003), Rinker et al. (1999), Yao, Huang, and Masten (1998). This reduction in the pH saves the cost of reducing the addition of acid for further treatment of Fenton process (Tizaoui et al. 2007). It was reported that the Ozonation-Fenton process was more efficient reported by Li et al. (2015) vol no:150 Journal of environmental Management.





Higher value of BOD5/COD obtained in the result indicates that Ozonation–Fenton process is more efficient than Fenton–Ozonation process. This was reported by many researchers that this is efficient in the treatment of landfill leachate for combined Ozonation–Fenton process. This was due to the synergic effect of ozone and Fenton reagent.

4. Conclusion

From the results, it was found that the influence of Microwave, O₃ and Fenton treatment on COD removal was effective. In Microwave 70% efficiency was achieved, efficiency up to 46% was achieved in O₃ treatment, 78.48% efficiency is achieved in O₃ and Fenton treatment. The microwave pretreatment significantly increased the COD removal with a short reaction time. This approach of combined Microwave, O₃ and Fenton treatment was more effective than Microwave or O₃ or Fenton treatment alone. Leachate subjected to O₃ and Fenton treatment resulted in reducing the reaction time with higher COD removal efficiency compared to the O₃ or Fenton treatment alone. Therefore, Microwave, O₃ and Fenton treatment provided a higher COD removal rate thus enhancing the leachate treatment.

Disclosure statement

No potential conflict of interest was reported by the author(s).

References

- Badawy, M. I., F. El-Gohary, T. A. Gad-Allah, and M. E. M. Ali. 2013. "Treatment of Landfill Leachate by Fenton Process: Parametric and Kinetic Studies." *Journal Desalination and Water Treatment* 51: 37–39.
- Boyle, W. C., and R. K. Ham. 1974. "Biological Treatability of Landfill Leachate." *Journal of Water Pollution Control Federation* 46 (5): 860–872.
- Cheng, S.-S., Y.-C. Chao, S.-C. Wong, C.-C. Chen, K.-H. Yang, and Y.-F. Yang. 2012. "Study on Hydrogen Production Potential Utilizing Leachate from Aerobic Bio-Leaching Bed Fed with Napiergrass and Kitchen Waste." *Energy Procedia* 72–81. doi:10.1016/j.egypro.2012.09.010.
- Cho, S. P., S. C. Hong, and S. Hong. 2002. "Photocatalytic Degradation of the Landfill Leachate Containing Refractory Matters and Nitrogen Compounds." *Applied Catalysis B: Environmental* 39: 125–133. doi:10.1016/S0926-3373(02)00079-6.
- Deng, Y., and J. D. Englehardt. 2006. "Treatment of Landfill Leachate by the Fenton Process." *Water Research* 40: 3683–3694. doi:10.1016/j.watres.2006.08.009.
- Deva, V. V., G. B. Raj, S. Antoya, V. Aruna, and K. A. Krishnan. 2020. "Zwitterion-chitosan Bed for the Simultaneous Immobilization of Zn(II), Cd(II), Pb(II) and Cu(II) from Multi-metal Aqueous Systems." *Journal of Cleaner Production* 255: 120309. doi:10.1016/j.jclepro.2020.120309.
- Goi, A., Y. Veressinina, and M. Trapido. 2009. "Combination of Ozonation and the Fenton Processes for Landfill Leachate Treatment: Evaluation of Treatment Efficiency." *Ozone Science and Engineering* 31 (1): 28–36. doi:10.1080/01919510802582011.
- Ho, S., W. C. Boyle, and R. K. Ham. 1974. "Chemical Treatment of Leachate from Sanitary Landfills." *Journal of Water Pollution Control Federation* 46 (7): 1776–1791.
- Kang, Y. W., and K. Y. Hwang. 2000. "Effects of Reaction Conditions on the Oxidation Efficiency in the Fenton Process." *Water Research* 34: 2786–2790.
- Kjeldsen, P., M. A. Barlaz, A. P. Rooker, A. Baun, A. Ledin, and T. H. Christensen. 2002. "Present and Long-term Composition of MSW Landfill Leachate: A Review." *Critical Reviews in Environmental Science and Technology* 32 (4): 297–336. doi:10.1080/10643380290813462.
- Krishnan, K. A., K. G. Sreejalekshmi, S. Varghese, and T. S. Anirudhan. 2010. "Removal of EDTA from Aqueous Solutions Using Activated Carbon Prepared from Rubber Wood Sawdust: Kinetic and Equilibrium Modeling." *Clean Soil Air Water* 38 (4): 361–369. doi:10.1002/clen.200900200.
- Krishnan, K. A., K. Ajmal, A. K. Faisal, and T. M. Liji. 2015a. "Kinetic and Isotherm Modeling of Methylene Blue Adsorption onto Kaolinite Clay at the Solid-Liquid Interface." *Separation Science and Technology* 50 (8): 1147–1157. doi:10.1080/01496395.2014.965832.
- Krishnan, K. A., S. S. Suresh, S. Arya, and K. G. S. Lekshmi. 2015b. "Adsorptive Removal of 2,4-dinitrophenol Using Active Carbon: Kinetic and Equilibrium Modeling at Solid-liquid Interface." *Desalination and Water Treatment* 54 (7): 1850–1861. doi:10.1080/19443994.2014.890548.
- Li, M., Z. Zeng, Y. Li, M. Arowo, J. Chen, H. Meng, and L. Shao. 2015. "Treatment of Amoxicillin by O₃/Fenton Process in a Rotating Packed Bed." *Journal of Environmental Management* 150: 404–411. doi:10.1016/j.jenvman.2014.12.019.
- Lin, H., H. Zhang, and L. Hou. 2014. "Degradation of C. I. Acid Orange 7 in Aqueous Solution by a Novel electro/Fe₃O₄/PDS Process." *Journal of Hazardous Materials* 276: 182–191. doi:10.1016/j.jhazmat.2014.05.021.
- Lopez, A., M. Pagano, A. Volpe, and A. C. Di Pinto. 2004. "Fenton's Pre-treatment of Mature Landfill Leachate." *Chemosphere* 54 (7): 1005–1010. doi:10.1016/j.chemosphere.2003.09.015.
- Nawrochi, J., J. Swietlik, U. Raczky-Stanislawiak, A. Dabrowska, S. Bilozor, and W. Ilecki. 2003. "Influence of Ozonation Conditionson Aldehyde and Carboxylic Acid Formation." *Ozone Science and Engineering* 25 (1): 53–62. doi:10.1080/713610650.
- Poblete, R. 2012. "Solar Photocatalytic Treatment of Landfill Leachate Using a Solid Mineral By-product as a Catalyst." *Chemosphere* 88: 1090–1096. doi:10.1016/j.chemosphere.2012.04.044.
- Poblete, R., E. Otal, L. F. Vilches, J. Vale, and C. Fernández-Pereira. 2011. "Photocatalytic Degradation of Humic

- Acids and Landfill Leachate Using a Solid Industrial By-product Containing TiO₂ and F⁻.” *Applied Catalysis B: Environmental* 102 (1–2): 172–179. doi:10.1016/j.apcatb.2010.11.039.
- Rinker, E. B., S. Ashour, M. C. Johnson, and G. J. Kott. 1999. “Kinetics of the Aqueous-phase Reaction between Ozone and 2,4,6-trichlorophenol.” *AIChE Journal* 45 (8): 1802–1807. doi:10.1002/aic.690450815.
- Rocha, E. M. R., V. J. P. Vilar, A. Fonseca, I. Saraiva, and R. A. R. Boaventura. 2011. “Landfill Leachate Treatment by Solar-driven AOPs.” *Solar Energy* 85: 46–56. doi:10.1016/j.solener.2010.11.001.
- Tizaoui, C., L. Bouselmi, L. Mansouri, and A. Ghrabi. 2007. “Landfill Leachate Treatment with Ozone and Ozone/hydrogen Peroxide Systems.” *Journal of Hazardous Material* 140: 316–324. doi:10.1016/j.jhazmat.2006.09.023.
- Yao, J.-J., Z.-H. Huang, and S. J. Masten. 1998. “The Ozonation of Benz[a]anthracene: Pathway and Product Identification.” *Water Research* 32 (11): 3235–3244. doi:10.1016/S0043-1354(98)00094-3.