

Treatment Processes of Leachate from a Landfill by Advanced Oxidation Fenton and Ozone-UV

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Abstract

In this paper it is presented the results of advanced oxidation of leachates from a technified sanitary landfill located in the State of Querétaro, Mexico. One characteristic of already stabilized leachates from sanitary landfills like this case, is their difficult degradation, mainly because the organic matter contained is recalcitrant. For the samples collect, four sites were selected, where three points per site were sampled, measuring at each site the parameters: temperature, pH, conductivity, redox potential (ORP) and dissolved oxygen (DO) and leachate samples were collected. On the other hand, the Chemical Oxygen Demand (COD) of crude leachates, leachates acidified and leachates oxidized by Fenton reagent and Ozone-UV combined were analyzed. COD was used to monitor the degradation kinetics. With the results, the ArcGIS software was applied to study the distribution of temperature, dissolved oxygen and COD mainly in the leachate lagoon. For the application of Fenton reagent in the crude leachate oxidation, the pH was first adjusted and Fe^{2+}/H_2O_2 ratio was optimized. The efficiency of Ozone-UV treatments was studied through COD degradation kinetics. The graphs of $\ln(C_i/Co)$ vs time, showed that the kinetic processes are of order one, with very acceptable regression coefficients (R^2) and extraordinarily similar speed constants (K). With Fenton oxidation, the highest percentage of COD degradation was achieved and with Ozone-UV oxidation, it was possible to practically degrade all the COD.

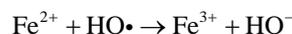
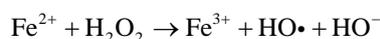
Keywords

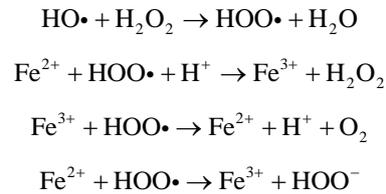
Landfill, Leachate, Fenton Oxidation, Ozone-UV Oxidation, Degradation

1. Introduction

In the construction of technical sanitary landfills, the land is selected with certain characteristics that allow to control the liquids (leachates) produced through oxidation ponds. A leachate is defined as a liquid that is formed by the reaction, dragging or filtering of the materials that constitute the waste and that contain in dissolved and suspended substances with high toxicity (SEMARNAT, 2003). The final disposal of Urban Solid Waste (MSW) has evolved from open-air landfills to highly technical landfills, where liquid (leachate) and potentially dangerous gaseous emissions for the environment are controlled in the latter. In both open-air landfills and early sanitary landfills, leachates had no control and these leached into surface water sources or infiltrated the soils, contaminating the aquifers underlying them. [1] [2]. The authors [3] [4] and [5] have considered that the phases in the decomposition of MSW in sanitary landfills are related to the age of the leachates. The author [6] made a classification of leachates based on their characteristics of pH, COD, TOC, BOD₅ and volatile fatty acids (VFA) and its age, in young (less than 5 years), medium (5 to 10 years) and old or stabilized (older than 10 years). The authors [6] and [7], studied and obtained leachate results, where the organic content was dependent on the class of decomposition conditions, the acetic acid and volatile fatty acids produced in the leachate in his study showed high values of COD and BOD₅. In the next degradation stage, these acids were converted to CO₂ and CH₄, so the organic content of the leachate decreased notably. Due to the content of leachates as organic components, their toxicity, its corrosiveness and their difficult degradation, it is important to establish some treatments for this type of substances that is easy to apply and, to a certain extent, economical. The difficult degradation, especially of stabilized leachates, can be through combined advanced oxidation treatments, such as the Fenton reaction as the first step, where a good number of organic components would be degraded, and to complete degradation it is possible to use an additional oxidation combining ozone with UV irradiation, since the problem of leachate lagoons in Mexico is that leachates of all ages are mixed.

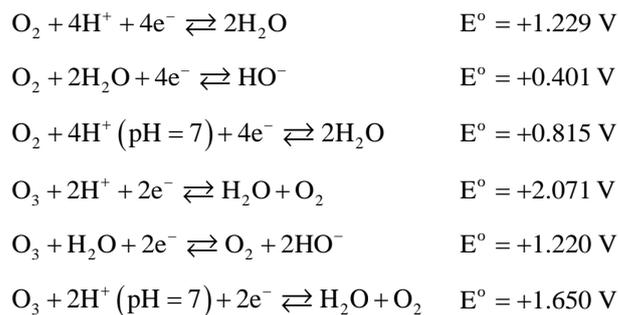
The oxidation by the Fenton reagent has been shown to be effective in the treatment of leachates [6] [8] [9] [10] and [11], in which the pollutant load is treated with a combination of Fe²⁺ and H₂O₂. This oxidation consists of a homogeneous catalytic system in which an iron salt, which can be FeSO₄ or FeCl₂, generates HO• radicals by the interaction of hydrogen peroxide (H₂O₂) with Fe²⁺. Author [12], proposed the following reactions:





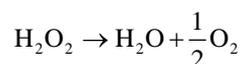
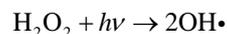
The range of applicability of the technique is also limited by the pH of the medium. The optimum pH for the reaction is between 3 and 6. With the Fenton oxidation, the degrees of reduction of the COD have reached up to 97% with respect to the initial values [13].

Regarding the treatment with ozone, its oxidizing power is strong, where its efficiency also depends on the pH, the presence of free radicals such as $\text{OH}\cdot$ and $\text{HOO}\cdot$ are active in the oxidation of pollutants. The following potentials illustrate the differences between the oxidative properties of ozone and oxygen [6] and [14]:

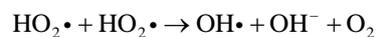
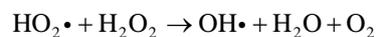
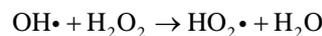


On the other hand, UV light is used in leachates treatment [15] is efficient in the presence of H_2O_2 , according to [16] and [17], it is based on the formation of hydroxyl radicals ($\text{OH}\cdot$) from the photolysis of hydrogen peroxide and the formation of radicals takes place due to the incident photons absorbed using lamps with an emission range between 180 and 400 nm, or more specific monochromatic (254 nm) to obtain a higher performance in the photolysis of peroxide. The main reactions of the oxidation mechanism by UV radiation are the following:

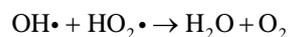
Initiation:



Propagation:



Termination:



The objective of this work is to present the results of a set of leachate treatments from the sanitary landfill of an Ecological Center for Integral Waste Management in the State of Querétaro (CEMIRQ, Mexico, through advanced oxidation by Fenton and subsequent treatment combined with ozone and UV.

2. Experimental Methodology

2.1. Sampling Site Location

The sample collection site was in a leachate lagoon of the sanitary landfill of the Ecological Center of Integral Waste Management Querétaro (CEMIRQ), in Querétaro State, Mexico. This is located near a cliff that blocks sunlight completely at two sites in the leachate lagoon, partially at one site and during the morning at another, **Figure 1**. The sampling was carried out considering the Mexico Standard [18] collecting three samples for each site as indicated in **Figure 1**. Each site was named: Site 1, Site 2, Site 3 and Site 4, a van Dorn equipment was used for sampling at 1.50 m depth; the lagoon has an average depth of 2.5 meters. These samples were stored at 4°C. The lagoon has a very dark color, **Figure 1**.

2.2. Determination of Physicochemical and Chemical Parameters

2.2.1. *In Situ* Parameters

The temperature, pH and conductivity were measured with a LabQuest-Vernier interface field equipment manufacturer's manual was considered for each case and the analysis were based on the Mexican standards [19] [20] and [21] respectively. Regarding the redox potential (ORP), a LabQuest-Vernier equipment and its respective electrode were also used, using the [22] ASTM technique, (1996). These parameters were measured three times for each of the three points at each site.

2.2.2. Analysis of Chemical Oxygen Demand (COD)

Organic matter was measured as COD. For analysis, the samples were oxidized in a HANNA Instruments HI839800 reactor, based on Standard Methods [23], It was subsequently measured in a DR/2400 HACH spectrophotometer, according to [23].



Figure 1. Sampling sites, three sampling points per site were considered.

2.2.3. Adjusting the pH, Optimization Fe/H₂O₂ Ratio and Fenton Oxidation

First the pH was adjusted between 3 - 5 with H₂SO₄ concentrated (J.T. Baker, 95.0% - 8.0%), this is really important for the correct addition of Fe²⁺ and H₂O₂. 5.11g of the iron salt (FeSO₄ 7H₂O, J.T. Baker 99.0% - 104.5%) were added and the H₂O₂ (J.T. Baker 30.0% - 32.0%) was slowly added until completing 15 mL to have the Fe²⁺/H₂O₂ ratio of 0.125 (previously optimized, following the [24] methodology, at a speed of 1 mL every minute, for one correct Fenton reaction treatment. The COD was utilized as a monitoring parameter.

2.2.4. Combined Ozone-UV Oxidation

The combined treatment with ozone and UV was carried out, after the Fenton oxidation with Batch technique. The parameter for ozonation and UV monitoring was also COD, this was monitored at different times: (0.20, 40, 60, 80, 100, 120, 140, 160, 180 min). The ozone was produced in a specifically built O3Residual brand generator, see **Figure 2(a)**. The ozone generator's duty cycle was established at 0.4 liters of oxygen per minute and an operating voltage of 7 kVolts. The ozonation reaction was carried out in a 31 × 3.8 cm quartz reactor with 34/45 standard mouth, for 8 hours, a pH range between 7 and 8. A Cole Parmer model lamp was used for UV radiation with high density ultraviolet light and long wavelength of 365 nm, intensity of 21,000 micro-W/CM² at 2" and 8900 micro-W/CM² at 10" that operates at 115 volts and 60 hz. In **Figure 2(b)**, the complete O₃-UV system is presented. All measurements and analysis of the different parameters were carried out in triplicate.

The leachate lagoon was mapped using ArcGIS software for the distribution of temperature, dissolved O₂ and COD at the four sites selected. The IDW and Arc Map interpolators were used to prepare the maps.

3. Results and Discussion

The results of this work are presented below.

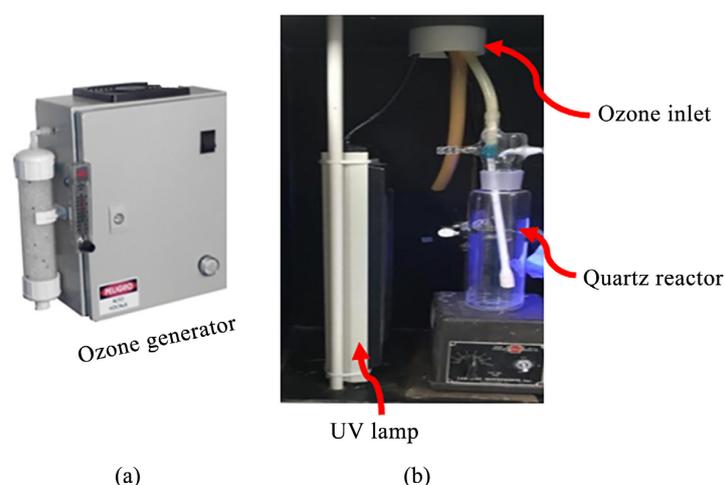


Figure 2. (a) Ozone generator. (b) Ozone-UV combined system.

3.1. Temperature

By analyzing the temperature, the highest values were obtained at the Sites 3 and 4, which are the sites that receive some sun and the lowest values were Site 2 and Site 1 that practically do not receive sun, due to a very high cliff that prevents the arrival of sunlight, **Figure 1**. In **Figure 3**, according to the ArcGIS software, it is shown how the temperature is distributed in the leachate lagoon, where the pink color presents the highest temperature values and the green color the lowest. It is noted that the temperature is not homogeneous in all lagoon, site 3 receives the most sunlight and sites 1 and 2 is the coldest.

3.2. pH of Leachates

The pH values were between 8.61 and 8.52, these values indicate that the leachate from the lagoon is stabilized. According to [7] and [24], those leachates with a pH greater than 8 are stabilized, have a low biodegradability $BOD_5/COD = 0.1$, $TOC/COD = 0.4$, indicating the presence of difficult oxidation compounds with weights greater than 5000 g/mol [6] and [25]. The pH is important in the composition of a leachate, because it depends on the concentrations of acids present and CO_2 that comes from the landfill. Rainfall and the waste type, are the most important factors affecting the composition of the leachate [26]. **Table 1** indicates the pH distribution in leachate lagoon, the Site 4 presented a somewhat higher pH, with respect to the other sites. These values are important, since all the leachate show stability, indicating the presence of compounds that are difficult to oxidize.

3.2.1. Conductivity

Table 1 shows the conductivity values, which were high, indicate the presence of high concentrations of dissolved ions. The magnitude of the pH suggests the presence of bicarbonates; by the smell and the almost zero OD concentration suggest that sulfides predominate.

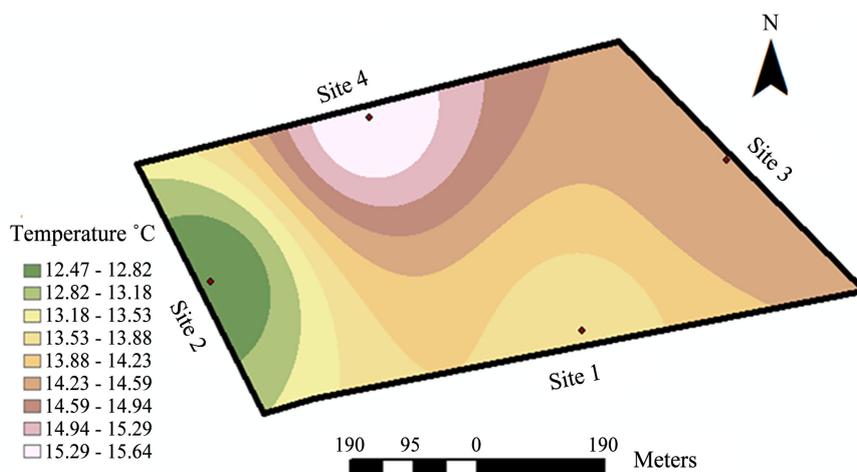


Figure 3. Temperature distribution according to ArcGIS software in the leachate lagoon.

Table 1. Values of the parameters measured in situ in the leachate lagoon.

Sites	Hydrogen potential (pH)	Conductivity ($\mu\text{S}/\text{cm}$)	Redox potential, ORP, (mV)
Site 1	8.51 ± 0.035	$34,400.00 \pm 547.723$	-194.40 ± 4.624
Site 2	8.55 ± 0.015	$33,522.67 \pm 517.178$	-197.78 ± 3.677
Site 3	8.55 ± 0.015	$40,977.00 \pm 112.892$	-203.96 ± 6.484
Site 4	8.59 ± 0.010	$39,485.33 \pm 264.652$	-203.67 ± 4.426

3.2.2. Redox Potential (ORP)

This parameter is closely related to DO. At the four sites very negative values were presented, which means very low oxygen concentration. This can be seen in **Table 1**. The sites that presented lowest negative values were Sites 3 and 4. This is consistent with temperature and solar incidence, since Site 4 receives most of the light.

3.3. Dissolved Oxygen (DO)

Figure 4 shows the Dissolved Oxygen (DO) distribution map, which together with the ORP are important parameters due to their relationship with COD. The importance of this parameter lies in that it indicates the degree of oxygenation of the leachate lagoon, which presents anoxia due to the very low concentration of DO at each site. These low values coincide with the negative values of the ORP. Since these leachates are already stabilized (this is presented by their dark color), it is logical that the DO concentration is very low, it can be seen in **Figure 2** that Site 4 shows a higher value, possibly due to the wind that allows a little dissolution of oxygen or perhaps the rain, although this oxygen is quickly used in the degradation of carbonaceous material. When analyzing the sites, it is observed that the leachate lagoon is acting as a practically anaerobic system.

3.4. COD from Crude Leachate

The most important parameter for leachate treatment is COD, **Figure 5** shows the distribution of COD in the leachate lagoon. Differences in COD values may be due to: the design of the sanitary landfill, the composition of the MSW, the characteristics of the site and the age of the sanitary landfill [6] and [7]. If the distribution map is analyzed, the COD presents an inverse panorama to that of DO since it is a parameter related to the amount of oxygen consumed by the oxidizable material in the lagoon, whatever its origin, organic or inorganic. COD, together with DO, are related to ORP (see **Table 1** for negative values).

For COD, its concentration in the crude leachate was first determined. **Table 2** shows the concentrations at each site, considering the analysis of the samples for every three points per site. **Figure 5** shows the distribution of COD in the leachate lagoon. Site 1 was the one with the highest concentration compared to the other sites, (green color). In general, the average crude leachate was around $14,940 \pm 85,987$. As indicated in the previous parameters, the COD was not homogeneous throughout the lagoon, Sites 3 and 4 presented less concentration which is also related to temperature, mainly where there is some solar radiation,

therefore, the process of oxidation of organic matter and the performance of anaerobic bacteria is greater.

3.5. COD from Leachate with Adjusted pH

To carry out the oxidation with the Fenton reagent, it was necessary to adjust the pH of each leachate to a value of 3 ± 0.144 , for which a volume of 0.009 ± 0.00045 mL of H_2SO_4 (concentrate) was used for each mL of crude leachate. **Table 2** shows the COD concentrations in mg/L, resulting after acidification. At this stage, when concentrated sulfuric acid was used, there was a small oxidation due to the presence of SO_3 , which is an oxide that works as a strong oxidant with

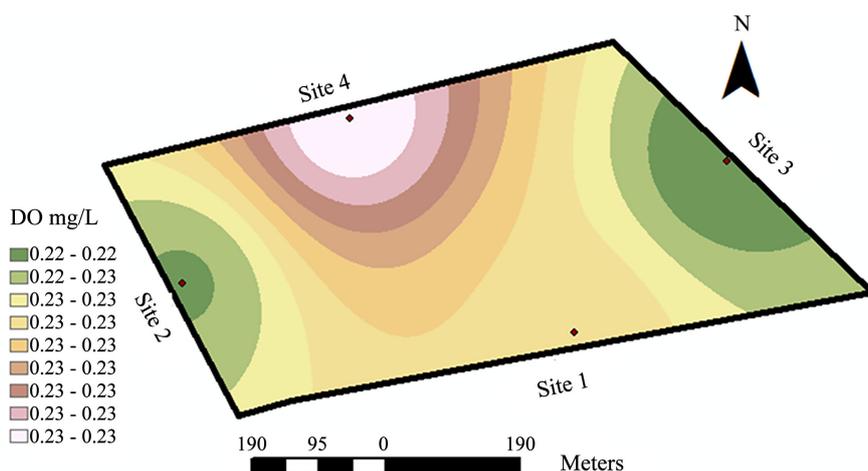


Figure 4. Oxygen dissolved distribution according to ArcGIS software in the leachate lagoon.

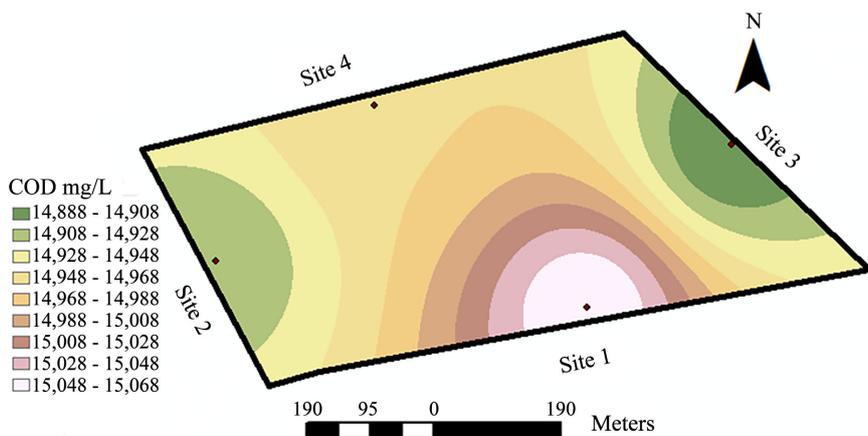


Figure 5. COD distribution according to ArcGIS software in the leachate lagoon.

Table 2. Stages of leachate treatment by Fenton oxidation.

Leachate	Site 1	Site 2	Site 3	Site 4
Crude (mg/L)	15,069.43 ± 19.84	14,904.82 ± 57.72	14,889.48 ± 29.97	14,912.04 ± 81.26
With adjusted pH (mg/L)	11,786.78 ± 65.72	11,716.55 ± 77.38	11,889.44 ± 57.67	11,764.83 ± 30.55
Oxidized by Fenton reagent (mg/L)	519.23 ± 15.66	671.35 ± 25.34	444.12 ± 30.54	454.53 ± 38.30

an oxidation potential of $E^\circ = +0.20$ V, thus helping in degrading the humic material. The reduction in COD of leachate per site was on average: for site 1: 3282.65 mg/L, for site 2: 3188.27 mg/L, for site 3: 3000.04 mg/L and for site 4: 3147.21 mg/L; If the averaged COD is considered in this lagoon of leachate, there was a reduction of 3154.54 mg/L which is equivalent to 21.11%.

3.6. Fenton Oxidation

The next step was to apply the Fenton reagent. **Table 2** shows the results of both sites and points of the leachate lagoon.

A strong decrease in COD was observed, due to the presence of the $\text{HO}\bullet$ and $\text{HOO}\bullet$ radicals. By considering average of the lagoon, there was a reduction of 14,421.63 mg/L, from beginning with the crude leachate, which is equivalent to 96.50%.

3.7. Complementary Oxidation Using an Ozone-UV Combination

Table 2 shows the residual leachate degradation values from the Fenton treatment, subsequently an oxidation was carried out by a combination of ozone and UV ($\text{O}_3 + \text{UV}$), producing a strong degradation. At this stage, the degradation kinetic curves were obtained constructing C_i/C_o graphs (C_i is the concentration over time and C_o is the initial concentration), to obtain the profile of the curve of each run, **Figure 6(a)**. By the shape of the curves, a first order kinetics was observed. That was corroborated when constructing the $\text{Ln } C_i/C_o$ vs Time curves. **Figure 6(b)** shows the logarithmic graphs $\text{Ln } C_i/C_o$ vs time of each run and their constants K , which were very close to each other.

3.8. Oxidation Treatments Representation and Performance Percentages in Each Stage

Finally in **Figure 7(a)** are presented how progressed the degradation of the leachate in each site of the lagoon in each treatment. From the crude leachate, after

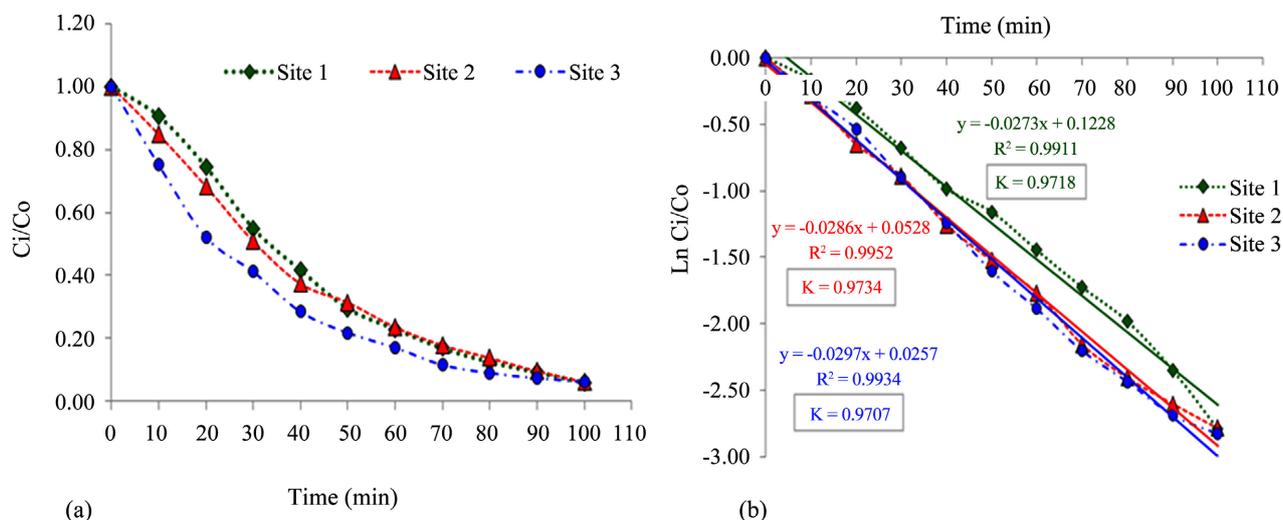


Figure 6. (a) Kinetics curves profile resulting from ozone-UV oxidation. (b) First order kinetic runs.

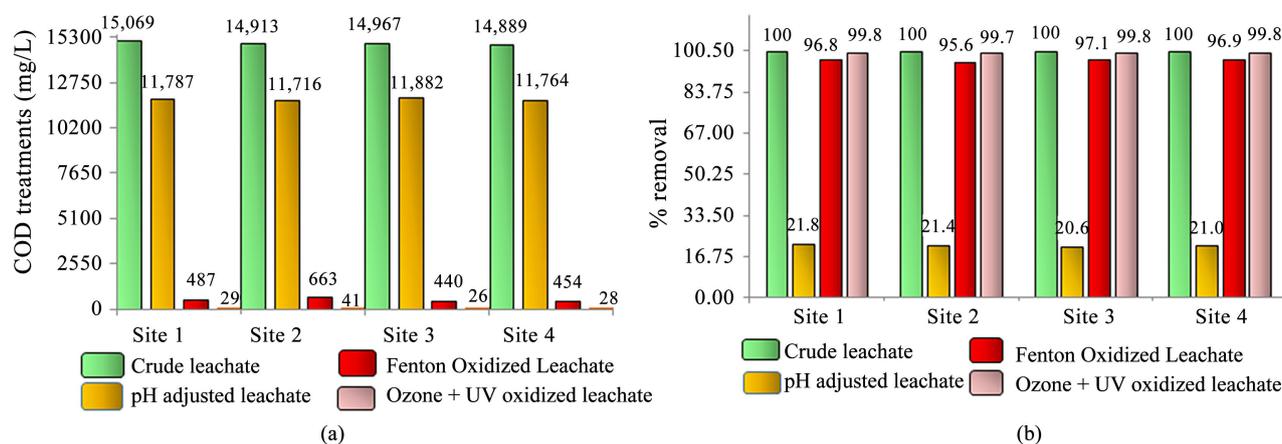


Figure 7. (a) Treatments stages. (b) COD removal efficiency in each treatment.

pH adjusted, where concentrated H_2SO_4 played an important role, at this stage the oxidation begins. Afterwards the Fenton reaction, which was the strongest and more efficient oxidation, there it was a great decrease in COD and finally with the ozone-UV combination oxidation, almost total degradation was achieved. By analyzing **Figure 7(b)**, the efficiency of each treatment is observed. Considering the combined ozone-UV oxidation treatment, there was a saving of time and the experimental error regarding the handling of the samples was minimized.

4. Conclusion

With the Fenton and the Ozone-UV Oxidations of these leachates, an efficiency greater than 96% was obtained, achieving complete the organic matter degradation. The pH adjustment was important. By means of the combined oxidation of the Ozone-UV system, the kinetic curves C_i/C_o vs time presented kinetics of the first order. The kinetic constants resulted of each run with values very close to each other. Performing the Ozone-UV combination in a single step allowed to save experimentation time and avoid experimental errors by handling the samples in less time than if it were done in separate steps.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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