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# The Performance of Microelectrolysis in Improving the Biodegradability Landfill Leachate

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**Abstract.** The aim of this study was to check the effectiveness of microelectrolysis for the pretreatment of a municipal landfill leachate with the objective improving its overall biodegradability, evaluated in terms of $BOD_5$/COD ratio, up to a value compatible with biological treatment. The best microelectrolysis operational conditions for achieving the desired COD values were: pH=2.0; granular activated carbon (GAC) =10 g/L; mass ratio of zero iron ($Fe^0$)/GAC=2:1; reaction time=90 min. The $BOD_5$/COD was significantly improved from 0.12 to 0.31, which allowed an almost 85% removal of COD by a sequential activated sludge process. The results show that the microelectrolysis is a promising technology to improve the biodegradability of mature landfill leachate.

## Introduction

Although landfill leachate has been proved to be toxic and recalcitrant, landfilling still remains one of the main methods for municipal and industrial solid waste disposal [1, 2]. Landfill leachate produced from sanitary landfills ought to be properly managed or treated. Otherwise, they can permeate ground water or mix with surface water [3, 4]. The composition and concentration of contaminants are influenced by seasonal weather variation, waste type, piling and mainly by the age of the landfill [5]. In general, the landfill leachate has higher $BOD_5$ values and even higher chemical oxygen demand (COD) contents [6]. However, mature landfill leachate characterized by a low $BOD_5$/COD ratio (<0.3) and a high fraction of high molecular weight organics. The biodegradation of the leachate is particularly challenging due to low biodegradable fraction of the organics and constituent toxicity to biological process [7, 8]. Hence, physicochemical processes may be required for pretreatment or full treatment of such leachate.

Traditional physicochemical processes, such as chemical precipitation, chemical oxidation, activated carbon adsorption, and advanced oxidation processes (AOPs) etc. have been proposed and practiced for the treatment of municipal landfill leachate. However, many of these technologies suffer the limitations of either being too expensive or not being able to treat mature landfill leachate.

Recently, microelectrolysis for treating poor biodegradable wastewater from pesticide factory [9], dyeing factory [10], and metal machining shop [11] has received wide attentions. From a chemical point of view, some authors [12, 13] suggested that organic contaminations can reacted with hydroxyl, atomic hydrogen and $Fe^{2+}$ produced during microelectrolysis. Additionally, organic contaminants can also be removed through adsorption, co-precipitation, and enmeshment in the ferrous and ferric hydroxide flocculation processes. Some authors [14] suggested that GAC can absorb some of the hydrophobic organic contaminants. From a biological point of view, other authors suggested that iron is an important and indispensable component of biomolecule which undergoes redox in cells [15], and $Fe^{2+}$ is often used as a coagulant in activated sludge treatment plants. Therefore, it could be economical and environmental to apply microelectrolysis to practical. With a sequential activated sludge process, physical, chemical and biological actions all take effects on removal of contaminants.

This study focused on the feasibility of microelectrolysis application for treatment of mature landfill leachate. A group of $L_9(3^4)$ orthogonal array experiments were used to find out the optimum conditions for treatment of mature landfill leachate by microelectrolysis. Reaction time, initial pH, dosage of GAC, mass ratio of $Fe^0$/GAC on reduction of organic constituents were tested through batch experiments. Then the biodegradability of between microelectrolysis pretreated leachate and raw leachate were compared by biodegradable process of leachate in laboratory scale.

## Experimental

**Landfill Leachate and Reagents.** The studied leachate was obtained from a sanitary landfill which was located in Guangzhou (Guangdong province, China) and closed in October 2002. It was collected in a zero headspace plastic bottle and refrigerated at 4℃ until use.

Sample of activated sludge inoculums were collected directly from the aeration tank of the municipal wastewater treatment plant. The sludge was continuously aerated using aeration pumps.

Iron scraps, Hydrogen peroxide ($H_2O_2$), Sulfuric ($H_2SO_4$) acid, Sodium hydroxide (NaOH) were purchased from a chemical reagent factory in Tianjin (China).

**Microelectrolysis Process.** In microelectrolysis process, after adjusting the lactate's pH to pre-set values by NaOH or $H_2SO_4$ solution, the lechate (100 ml) and pre-set dosages of GAC and iron scraps were added into 500 ml beaker. A period time latter, supernatant was taken out to monitor the COD and pH. An orthogonal array experiment $L_9(3^4)$ was designed to determine the optimum microelectrolytic conditions for the removal of COD from landfill leachate solution. Four significant factors were selected based on the experimental of other authors [11, 16]. Then, batch experiments were taken to investigate the effect of dosage on the removal of COD from aqueous solution. All experiments were conducted in duplicate and the averaged data were reported.

**Biological Procedure.** The activated sludge system was applied in cubical aeration glass-vessels with total volume of about 1200 ml. The system was aerated by using air pumps. The initial volume of the culture was 500 ml, which was completed to 1000 ml with substrates (leachate, pretreated leachate or glucose) at the beginning of each cycle. All the experiments were carried out in duplicate and at room temperature (21-26℃) by periods of 72 h. Physical characteristics of the sludge were periodically and microscopic observation. For COD determinations, samples (25 ml each) were taken every 12 h, after they had been filtered through a 0.45μm Millipore filter.

## Results and Discussion

**Characterization of The mature Landfill Leachate.** The main chemical characteristics of landfill leachate were summarized in table 1. With biodegradability ratio ($BOD_5$/COD) lower than 0.13 and pH higher than 8, the sample can normally classified as refractory to conventional biodegradation processes [6, 16]. Physicochemical processes are necessaries for the treatment of mature leachate.

Table 1. Chemical characteristics of the studied landfill leachate

| Parameters | Values |
|---|---|
| pH | 8.5±0.1 |
| Color | 1500±50 |
| COD | 3318.2±205.5 |
| $BOD_5$ | 397.5±48.5 |
| TOC | 843.4±71.5 |
| SS | 330.4±47.5 |

**Optimization of Microelectrolysis Process.** Levels of each operating factor and the design matrix of the $L_9(3^4)$ orthogonal array were summarized in Table 2. Characteristic values used in this experimental design were the overall COD removal efficiency and pH. The experiments were conducted three times each time to ensure the accuracy of the experimental data.

The ANOVA results for the experiment are tabulated in Table 3. On the basis of the calculated F-values, the dosage of GAC, reaction time, and initial pH are statistically inferred to have significant influences on COD removal in treatment of mature landfill leachate. Their order of influence in terms of COD removal efficiency was dosage of GAC > reaction time >initial pH. According to the results of Table 2 and Table 3, in the following experiments, the selected dosage of GAC was 10 g/L, initial pH was adjusted to 2, reaction time was about 90 min, and mass ratio of $Fe^0$/GAC was 2:1.

Table 2. Design matrix and experimental results for the $L_9(3^4)$ orthogonal array experiments

| Experiment | Operating factors and their levels | | | | Mean responses | | |
|---|---|---|---|---|---|---|---|
| | ($Fe^0$/GAC ratio) | GAC (mg/L) | Initial pH | Reaction time (min) | Overall COD removal (%) | COD(mg/L) | pH |
| 1 | 1:2 | 1 | 2 | 30 | 44.5 | 1836.7 | 2.1 |
| 2 | 1:2 | 5 | 3 | 60 | 41.5 | 1938.1 | 5.7 |
| 3 | 1:2 | 10 | 4 | 90 | 73.1 | 890.7 | 7.6 |
| 4 | 1:1 | 1 | 3 | 90 | 40.5 | 1971.1 | 3.4 |
| 5 | 1:1 | 5 | 4 | 30 | 50.3 | 1645.4 | 6.4 |
| 6 | 1:1 | 10 | 2 | 60 | 77.3 | 750.5 | 3.0 |
| 7 | 2:1 | 1 | 4 | 60 | 44.0 | 1855.7 | 5.0 |
| 8 | 2:1 | 5 | 2 | 90 | 65.1 | 1154.6 | 2.5 |
| 9 | 2:1 | 10 | 3 | 30 | 73.4 | 882.5 | 6.8 |

Table 3. Analysis of variance (ANOVA) for COD removal efficiencies in the $L_9(3^4)$ orthogonal array experiments

| Operating factor | Degrees of freedom (d.f) | Sum of Squares (SS) | Mean square (MS) | F ratio |
|---|---|---|---|---|
| COD removal | | | | |
| $Fe^0$/GAC ratio | 2 | 77.9 | 39.0 | 1.2 |
| GAC (mg/L) | 2 | 4077.9 | 2039.0 | 62.1 |
| Initial pH | 2 | 202.5 | 101.3 | 3.1 |
| Reaction time (min) | 2 | 280.1 | 140.1 | 4.3 |
| Error | 18 | 590.7 | 32.8 | |
| Total | 26 | | | |

Significance level at a 95% confidence interval, $F(2, 18)_{0.05}=3.0$

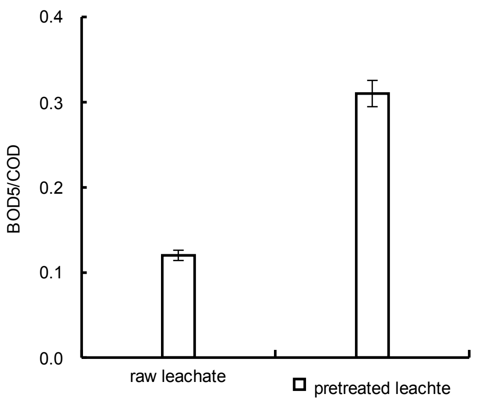


Fig.1. The BOD5/COD change after pretreated by microelectrolysis.

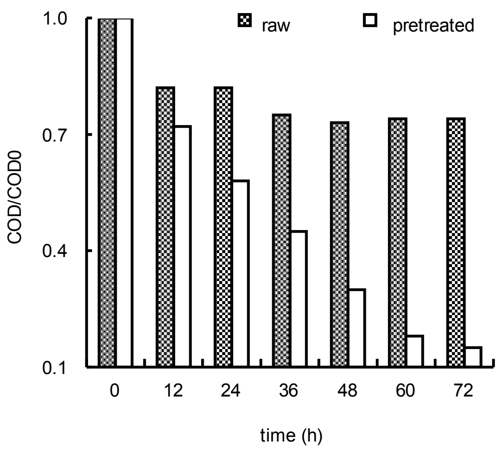


Fig.2. Evolution of COD during biological treatment of the leachate.

**Biodegradability Changes during Microelectrolysis Decomposition.** The biodegradability of the leachate was evaluated through the evolution of the $BOD_5$/COD ratio. Fig. 1 shows that for untreated samples, this parameter attains values of about 0.12 while microelectrolysis treatment permit its enhancement up to 0.31, which represent substantial biodegradability according to the current literature [17]. This is a fact of remarkable importance in the case of the application of microelectrolysis-biological integrated system to wastewater treatment. It is admitted that microelectrolysis can improving the efficiency and reducing the cost of further biological process.

In a second phase, raw and pretreated leachate was submitted to a biological degradation process using a sequential batch reactor. The evolution of COD during the biological treatment (Fig.2) conform the low biodegradability of raw mature leachate, which achieves a maximal COD removal of about 25% at a 72 h treatment time. On the other hand, the COD of microelectrolysis pretreated leachate fades progressively attaining COD removal higher than 85% at the end of the 72 h. This result indicated that the microelectrolysis process can get rid of some non-biodegradable organics or convert the non-biodegradable organics to more biodegradable forms.

## Conclusion

An investigation aimed at checking the effectiveness of microelectrolysis for the enhancement of mature landfill leachate has been carried out at lab scale at ambient temperature. Under the most optimal microelectrolysis operational conditions, 75-80% COD can be moved effectively. The removal of COD was influenced by the dosage of GAC, reaction time, initial pH and mass ratio of $Fe^0$/GAC. Their order of influence in terms of COD removal efficiency was dosage of GAC > reaction time >initial pH > $Fe^0$/GAC.

The biological degradation experiment demonstrate that microelectrolysis can transform organic recalcitrant compounds into easily biodegradable products or can get rid of some non-biodegradable organics, improving the efficiency and reducing the cost of further biological process.

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