DECOLORIZATION OF LANDFILL LEACHATE USING ELECTROCHEMICAL TECHNIQUE

ZAINAB HAIDER MUSSA[a*], MOHAMED ROZALI OTHMAN[a], Md PUAZI ABDULLAH[a,b] and NORAZZIZI NORDIN[a]

[a]School of Chemical Sciences and Food Technology, Faculty of Sciences and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, MALAYSIA
[b]Centre for Water Research and Analysis (ALIR), Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, MALAYSIA

ABSTRACT

Several operating conditions such as electrode material, treatment time, applied voltage and Cl^- concentration were tested on the treatment landfill leachate using electrochemical (EC) method. Results obtained show that EC method can be used for treatment of landfill leachate by using proper operating condition. The best removal rates were obtained when graphite rod electrode was used as an anode, operating time is 120 min, voltage applied is 10 V, NaCl concentration is 0.585 (w/v), 80% of color, 72% of COD, 45% of NH_3-N, 63% of Total–P (PO_4^{-3}) removal were obtained.

Key words: Landfill leachate, Jeram, Decolorization, Electrochemical.

INTRODUCTION

In landfill, solid waste undergoes physico-chemical and biological changes. Digestion of the organic fraction of the wastes in combination with percolating rainwater produces a highly contaminated liquid called leachate, which is the most important point source of organic ground water contamination[1]. Leachate can be categorized as a liquid waste that contains high chemical oxygen demand (COD), high levels of ammonia and phosphorus[2]. It can readily pollute soil and penetrates into the underground layers of ground resulting in severe underground water contamination which is one of the major water sources for human societies[3]. Landfill leachate becomes an issue as a wastewater sources since it may cause serious pollution to ecosystem. In order to reach environment friendly criteria for landfill leachate, one must bring these values to an acceptable discharge limit. Hence, landfill leachate must be collected and treated. Therefore, many pretreatment and combined

---

*Author for correspondence; E-mail: zp69014@yahoo.com; Ph.: +60172568779

treatment methods have been proven to treat leachate such as biological treatment methods[4], membrane processes[5,6] advanced oxidation techniques[7,8], coagulation-flocculation methods[9], lagoon and wetland application[10]. Since the variation in volume and composition, these methods have troubles such as decreasing treatment efficiencies and increasing cost[7]. Therefore, more effective treatment methods have been proved to treat leachate. In recent years, electrochemical oxidation process has been shown to be promising for landfill leachate treatment[11]. It is an alternative technology for water and wastewater treatment systems and most effective in removing inorganic and organic contaminants. In this technique, which is characterized by its simple equipment, easy to operate and automation and no chemical requirement. Electrochemical treatment processes have been used for long time to remove not only particulate materials but also treat both COD and NH_3-N simultaneously and is capable of providing a high color removal[11]. In general, pollutants can be destroyed electro-chemically by direct anodic oxidation or by indirect oxidation[12]. The aim of this study is to evaluate the effectiveness of electrochemical oxidation for leachate treatment and determine the optimum operational conditions (voltage applied, Cl^- concentration and reaction time) for leachate treatment.

EXPERIMENTAL

Sampling

Leachate samples were collected monthly from February to July in 2013. Samples of leachate were collected from Jeram Sanitary Landfill, which is located in an oil palm plantation near Mukim Jeram, Kuala Selangor. It is roughly rectangle in shape and occupied 64.7 hectares. Jeram sanitary landfill has been in operation by Worldwide Holdings since January 2007. Approximately 1,500 tons of waste per day was received in this site and it had processed about 3.0 million of municipal solid waste. To investigate the physico-chemical parameters of raw leachate, real sample was analyzed to evaluate its initial COD, NH_3-N, pH, color, conductivity, TDS, and Total–P (PO_4^{-3}) as presented in Table 1.

Table 1: The characteristics of leachate

Parameter	Data of sampling				
	February	**March**	**April**	**June**	**July**
COD mg/L	10000	6250	8000	10250	11250
NH_3-N mg/L	5800	9800	6400	7600	2800

Parameter	Data of sampling				
	February	**March**	**April**	**June**	**July**
Color Pt-Co	11500	12900	9100	20800	24300
Total-P mg/L	126	117	210	210	135
TDS ppt	26.9	29.5	25.9	23.9	19.39
pH	8.17	8.5	7.6	8.4	8.28
Conductivity mS/cm	29.2	30.5	28.1	25.9	21.05

Reagents

All regents were used as received without further purification. The chemical oxygen demand reagent (COD) Cat.21258-25 and Total-P (PO_4^{-3}) reagent (Cat. 27427-45) were provided by (Loveland, U.S.A.), NH_3-N reagent (Cat.26531-99, Cat.26532-99) was provided by (Dusseldorf, Germany). Hydrochloric acid, sodium chloride and sodium hydroxide were purchased from Merck with purities of more than 99.5%.

The reactor

Laboratory scale electrolysis cell system was used as an electrochemical reactor, it is consisted of a DC power supply and a glass reactor (100 mL volume) with 1 cm × 1 cm platinum plate as cathode and graphite rod as anode. The distance between the electrodes was approximately 2 cm. The electrodes were connected to the DC power supply (CPX200 DUAL, 35V 10A PSU). Schematic representation of the rector is presented in Fig. 1.
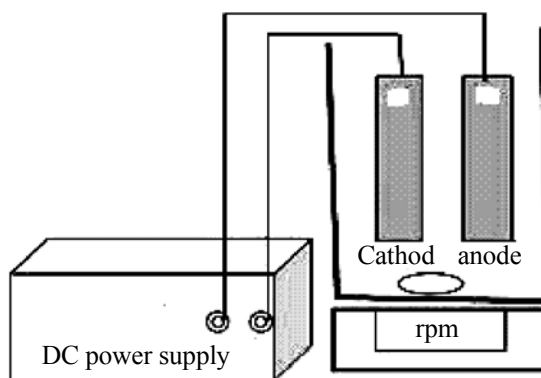

Fig. 1: The schematic experimental setup in this study

Preparation of electrodes

Ni, Ag, Cu, Pd and Pt metal foils were used to prepare Ni, Ag, Cu, Pd and Pt electrodes, respectively. A 0.5 mm thick Ni, Ag, Cu, Pd and Pt foils were cut into approximately 1 cm × 1 cm piece and conducting to silver wire with silver conducting paint prior covered with epoxy gum.

Treatment process

All experiments were performed by using the laboratory electrolysis cell which is consisted of a DC power supply, and a glass electrochemical reactor, 40 mL of landfill leachate and a known amount of supporting electrolyte NaCl was added to leachate. During each experiment, mixing of the rector contents was maintained to keep the solution homogenous using magnetic stirrer. All experiments were carried out at temperature controlled between 24 to 25 by circulating water bath. Treated samples were taken at regular time intervals. Before each experiment, electrodes were polished using sand papers to remove surface grease then washed by distilled water and acetone. The operational conditions of the experiments are given in Table 2. Leachate color, COD, NH_3-N and total phosphorus PO_4^{-3} were measured by a Hach Odyssey DR 2400 spectrophotometer and a Hach COD reactor. The pH was measured by a pH meter. TDS and conductivity were measured by using conductivity meter (cond 610). Using this method, the removal efficiency (%R) of landfill leachate can be obtained at any time, with respect to its initial values. The energy consumption (Esp) was calculated during the process according to Eq. (1)[13].

$$E_{sp} = \frac{E\,I\,t}{3600\,V} \qquad \qquad ...(1)$$

Where E_{sp}, energy consmption in Wh/L, E is the applied voltage, I is the current intensity, in A, t is the electrocoagulation time, in s, V is the volume, of the sample, in L.

The removal percentage for COD, Color, NH_3 and Total-P was calculated according to Eq. (2)[14].

$$R\,\% = \frac{A_0 - A_t}{A_0}\,x\,100 \qquad \qquad ...(2)$$

Where R% is the removal percentage for parameters (COD, Color, NH_3-N and Total-P), A_0 is initial value of parameters, A_t is the value of parameters at time t.

Table 2: The operative conditions

Parameter	Value
Landfill leachate	40 mL
Electrolyte amount (w/v)	0.05, 0.15, 0.375 and 0.585
Applied voltage (V)	5, 10, 15 and 20
Temperature (°C)	Ambient (lab scale)
Time (min)	30, 60 and 120

RESULTS AND DISCUSION

The effect of electrode materials

Different electrode materials affect on the performance of the electrochemical process. In this study totally 7 electrodes materials consisting of platinum (Pt), palladium (Pd), nickel (Ni), silver (Ag), copper (Cu) plates, graphite rod and stainsteel rod as anode were optimized in terms of color removal on the condition that 10 voltage, 120 min electrolysis time, the raw PH (pH = 8.3) and NaCl as electrolyte supporting o.585 (w/v), as can be seen in Fig. 2.
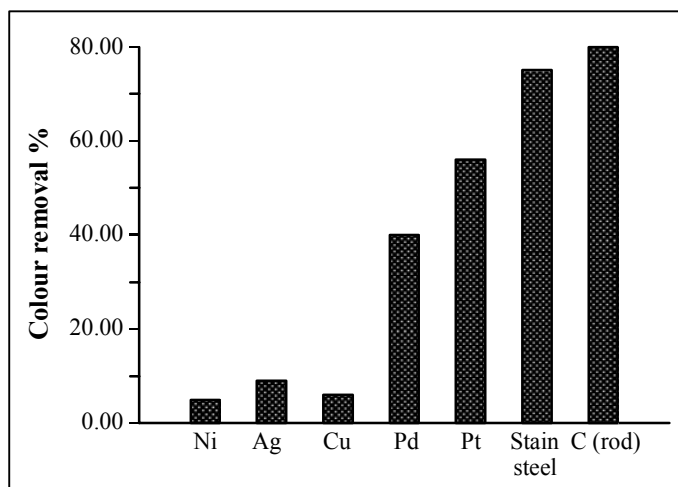

Fig. 2: Optimization of several electrode based on color removal

Good color removal (80%) was observed during 120 min using graphite rod compared with others. Meanwhile, the result of color removal using Ni, Ag, Cu, Pd, Pt, and

stainsteel were 5, 9, 6, 40, 56 and 75%, respectively. On the other hand, the difference between the rate of removal (% color removal) for graphite rod and stainsteel is just about (5%), so the choosing of best electrode was made not only based on their decolorization ability but also based on their stability electrochemically, as shown in Table 3. Graphite rod electrode was selected for further experiments.

Table 3: Electrochemical oxidation of landfill leachate in 0.585 (w/v) NaCl using various electrodes as an anode and platinum (Pt) as cathode. Voltage of 10 V was applied

Anode materials	Decolorization time (min)	Decolorization percentage (%)	Observation on	
			Electrolysis product	**Anode**
Nickel (Ni)	120	5	Color unchange and precipitation	Completely corroded
Silver (Ag)	120	9	Color unchanged	Slightly corroded
Copper (Cu)	120	6	Color unchange and precipitation	Completely corroded
Palladium (Pd)	120	40	Yellowish solution	Unchange
Platinum (Pt)	120	56	Yellowish solution	Unchange
Stainsteel	120	75	Yellowish solution and precipitation	Slightly corroded
Graphite rod	120	80	Yellowish solution	Unchange

Investigation the effect of applied voltage

The effect of voltage on color removal

One of the most important parameters that can affect the removal in the electrochemical process is the applied current or the voltage. Color removal efficiencies were significantly affected by the cell voltage. In order to determine the efficiency of color removal from landfill leachate, different electrical potential (voltage) values were used. The results are presented in Fig. 3. At any time during electrolysis increasing the applied voltage was accompanied by an increase in the percentage of color removal. The maximum color removal rate was attained at the potential voltage of 20 V at treatment time of 120 min. Color removal rates for 5, 10 and 15 V were determined to be 20, 80 and 87% respectively, for the 120 min treatment time. This behavior may be due to the increased generation of OCl^{-1}

species[15]. Moreover, the rate of bubble-generation increases and the bubble size decreases with the increasing of voltage, resulting in a faster removal of pollutants by H_2 flotation[16].
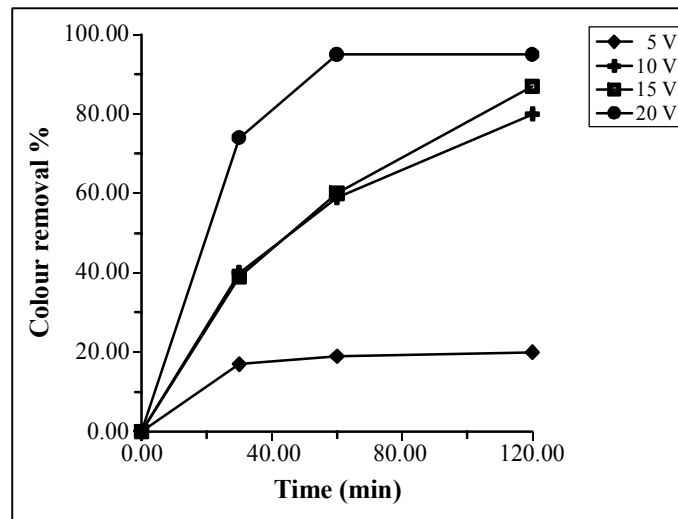

Fig. 3: Effect of applied voltage on color removal

Determination of the optimum cell voltage based on energy consumption (E_{sp})

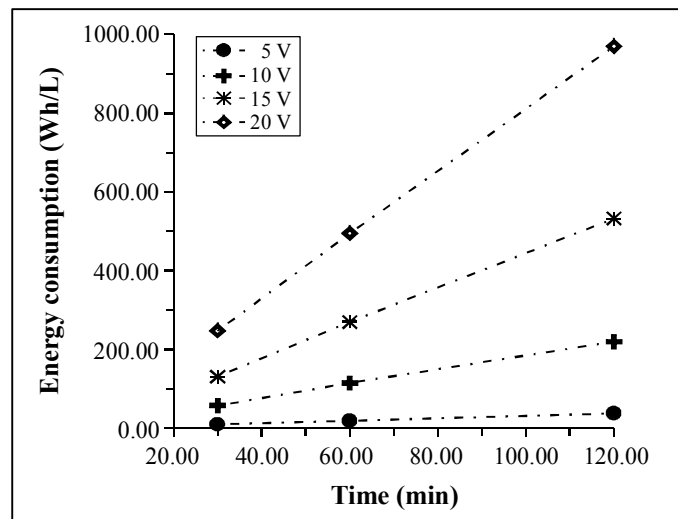The energy consumption as a function of applied voltage was calculated and the results are shown in Fig. 4.

Fig. 4: Effect of voltage on consumption energy

It is clear from the figure that (E_{sp}) increase with voltage, when potential voltage was applied as 5 V, unit energy consumption were obtained as 37.5 Wh/L treated leachate. When voltage was applied as 10 V, unit energy consumption were obtained 220 Wh/L treated leachate. When voltage was applied 15 V, unit energy consumption was obtained 532.5 Wh/L treated leachate and at 20V, unit energy consumption was 970 Wh/L. These results showed that 10 V as the optimal electrical potential for the EC process. Because it ensures the removal rate accompanied with reducing the electrical energy to reach desired color removal, Therefor, 10 V of applied voltage has been selected for the further experiments.

Effect of electrolysis time on COD, Color, NH_3-N and Total-P (PO_4^{-3})

The effect of electrolysis time was investigated in the range 30 min to 120 min by the following condition; raw pH, 10V, values. As can be seen in Fig. 5.
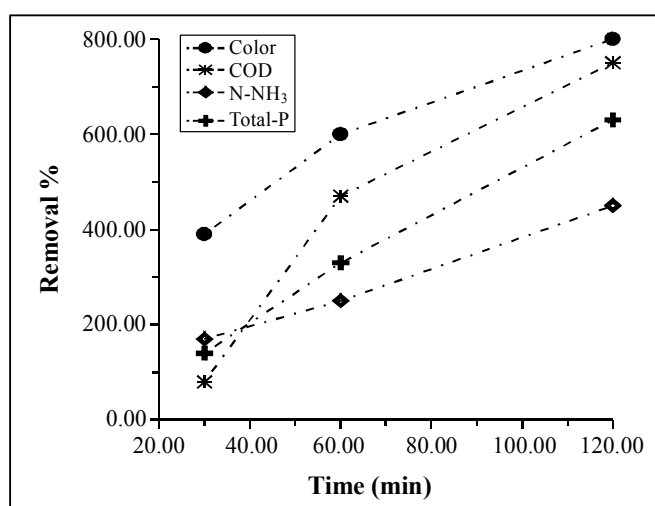

Fig. 5: Effect of electrolysis time on color, COD, NH_3-N and total-P (PO_4^{-3}) removal

An increase in the time from 30 to 120 minutes yield an increase in the efficiency of COD removal from 8% to 72%, color removal from 39% to 80%, NH_3-N from 17% to 45% and total-P (PO_4^{-3}) from 14% to 63%. The reason that, chloride ions usually present in landfill leachates are oxidized to chlorine/hypochlorite. All of these species are responsible for the oxidation of organic and inorganic pollutants[17]. That is mean, organic and inorganic compounds removal efficiency depends on the concentration of hypochlorite ion that generated electrochemically in solution. When the electrolysis time was longer more hypochlorite produces in solution under fixed voltage, thus COD, NH_3-N. Color, total phosphorous concentration in the solution were reduced in higher concentration of hypochlorite.

The role of supporting electrolyte

The electrical conductivity of the solution is an important parameter for saving electric energy[18]. To increases the electric conductivity of the solution, NaCl was used as the supporting electrolyte. The time is 30 min, 60 min, 120 min and potential voltage is 10 V. The effect of Cl^- on the removal efficiency was studied at 0.05, 0.15, 0.375 and 0.585 (w/v) levels, as can be seen in Fig. 6a and 6b.
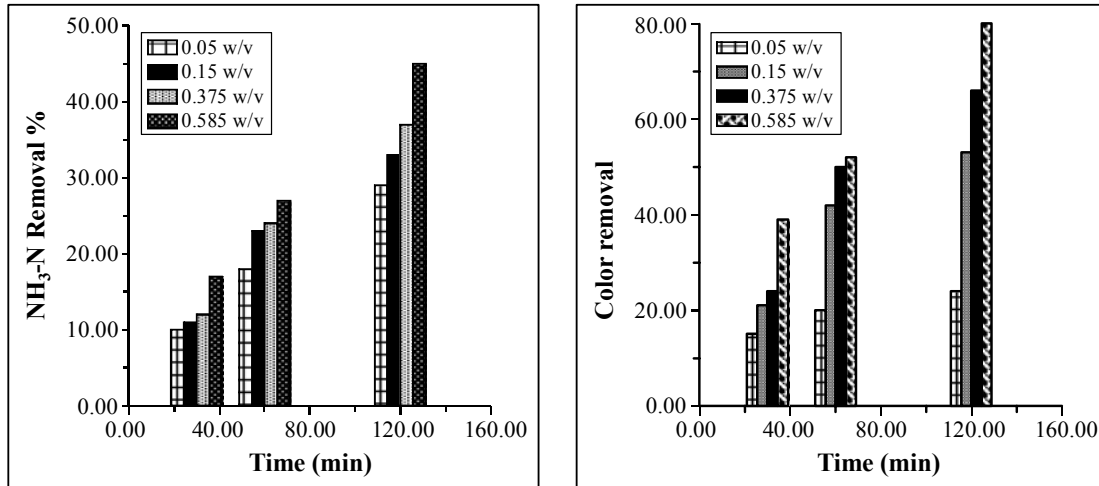

Fig. 6a: Effect of Cl^- concentration on NH_3-N removal and color
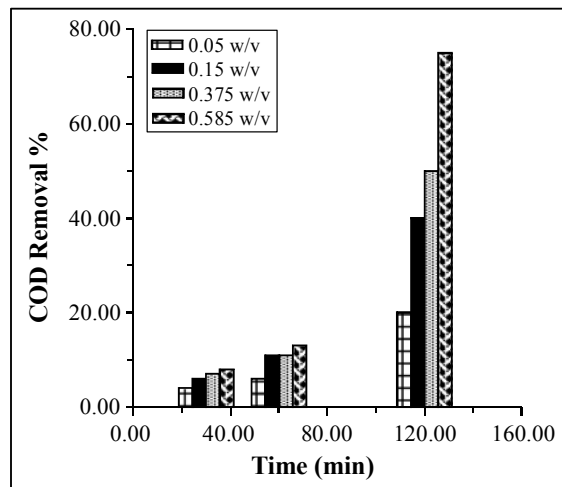

Fig. 6b: Effect of Cl^- concentration on COD removal

An increase in the concentration of Cl^- from 0.05 to 0.585 (w/v) yield an increase in the efficiency of COD removal from 27% to 72%, color removal from 40% to 80%, NH_3-N from 30% to 45% and total-P (PO_4^{-3}) from 14% to 63%. During the coagulation process, the Cl^- will be discharged at the anode to generate Cl_2, then the Cl_2 can be chemically convert to ClO^- which is able to oxidize the pollutants[19].

CONCLUSION

The present investigation illustrates that the most suitable conditions for treatment landfill leachate are graphite electrode as anode, supporting electrolyte; NaCl with concentration 0.585 (w/v), applied voltage of 10 V and electrolysis time is 120 min. The obtained results confirm that EC method can be used efficiently for the treatment of landfill leachate.

ACKNOWLEDGEMENT

REFRENCES

1.	T. Amir, J. Sustainable Development, **2,** 159 (2009).

2.	M. F. Niam, F. Othman, J. Sohaili and Z. Fauzia, Water Sci. Tecnol., **56**, 47 (2007).

3.	S. Renou, J. G. Givaudan, S. Polulain, F. Dirassouyan and P. Moulin, J. Hazard. Mater., **150**, 468 (2008).

4.	U. Welander, T. Henrysson and T. Welander, Water Res., **32,** 1564 (1998).

5.	A. Chianese, R. Ranauro and N. Verdone, Water Res., **33,** 647 (1999).

6.	K. Tabet, Ph. Moulin, J. D. Vilomet, A. Amberto and F. Charbit, Separat. Sci. Technol., **37**, 1041 (2002).

7.	J. J. Wu, C. C. Wu, H. W. Ma and C. C. Chang, Chemosphere, **54**, 997 (2004).

8.	H. Zhang, D. Zhang and J. Zhou, J. Hazard. Mater., **135**, 106 (2006).

9.	A. Amokrane, C. Comel and J. Veron, Water Res., **31**, 2775 (1997).

10.	T. Maehlum, Water Sci. Technol., **32**, 129 (1995).

11.	F. IIhan, U. Kurt, O. Apaydin and M. T. Gonullu, J. Hazard. Mater., **154**, 381 (2008).

12. C. C. Li, J. E. Chang and T. W. Chin, Water Res., **29**, 671 (1995).

13. D. Norma, A. Fernandes, L. Ciriaco J. Pacheco and A. Lopes, Purtugaliae Electrochimica Acta, **30**, 281 (2012).

14. M. Panizza and C. A. Martinez-Huitle, Chemosphere, **90**, 1455 (2013).

15. J. B. Parsa, M. Rezaei and A. R. Soleymani, J. Hazard., **168**, 997 (2009).

16. O. T. Can and M. Bayramoglu, J. Hazard Mater., **173**, 731 (2010).

17. X. Zhao, J. Qu, H. Liu, C. Wang, S. Xiao, R. Liu, P. Liu, H. Lan and C. Hu, Bioresource Technology, **101**, 865 (2010)

18. W. L. Chou, C. T. Wang and S. Y. Chang, J. Hazard. Mater., **186**, 1200 (2009).

19. C. T. Wang, W. L. Chou and Y. M. Kuo, J. Hazard. Mater., **164,** 81 (2009).