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FABRICATION OF SELECTED METAL POWDER COMPOSITE ELECTRODE FOR LANDFILL LEACHATE TREATMENT USING ELECTROCHEMICAL METHOD

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ABSTRACT

Landfill leachate is a major source of pollution caused by the wastewater generated from solid waste buried underground. Electrochemical oxidation processes, is one of the environmental friendly technologies in landfill leachate treatment processes. In this research, several composite electrodes of metal powder (Fe, Pt, Ni, Co, Cu, Al, Ag) with PVC were used by mixing together a weighed portion of metal powder and PVC in THF as a solvent. The mixture were then swirled until the suspension was homogeneous and drying the suspension in an oven at 100°C for 3 hr. The dry sample was then placed in a 1 cm diameter stainless steel mould and pressed at 10 ton/cm^2. The ready composite electrodes were then used as a working electrode to treat a leachate samples using electrolysis method with stainless steel as the counter electrode. The results showed a composition of Pt_{85}-PVC_{15} was the best electrode, which gavethe removal percentage of color, COD and NH_3-N are 99, 98 and 91%, respectively. The optimum operating conditions were sodium chloride concentration of 1.2% (w/v) as a supporting electrolyte, applied voltage of 10 V and operating time of 90 min.

Key words: Landfill leachate, Fabrication, Electrode, Electrochemical.

INTRODUCTION

Sanitary landfills are one of the most commonly used methods for the management of solid wastes produced by the municipal councils or cities around the world[1]. Growing trend in population, urbanization and industrialization has resulted high amount of solid wastes being generated. The generation of municipal solid waste has increased day by day due to rapid development of urban areas and rural-urban migration. Landfill leachate contains high concentration of pollutants include chemical oxygen demand (COD), ammonia,

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phosphorus as well as total dissolved solid and becoming an issue as a wastewater sources, since it may cause serious pollution to ecosystem[2]. Humid substances represent > 60% of the total organic C found in anaerobic landfill leachates. In an effort to control the pollution caused by landfill leachate, many treatment processes have been studied, including biological treatment[3], photocatalysis[4], adsorption[5], chemical precipitation[6], Fenton's oxidative treatment[7], coagulation/flocculation[8,9], advanced oxidation processes[1,10] and membrane processes[11,12]. These test methods have troubles such as decreasing treatment efficiencies and increasing cost[1]. Upto now, electrochemical oxidation process has been proved to be promising for wastewater treatment mainly due to its high effectiveness, easy to operate and more economic. Especially in the case of landfill leachate, which has rich chloride ions and good conductivity. Electrochemical oxidation technique is an effective method not only for color removal but also for COD removal. The pollutants are destroyed by either direct or indirect oxidation process. The main aim of this study is to fabricate metal powder electrode to treat landfill leachate and determine the optimum operational conditions (reaction time, voltage applied, and Cl^- concentration).

EXPERIMENTAL

Sampling

Leachate samples were collected from Jeram Sanitary Landfill, which is located in an oil palm plantation near Mukim Jeram, Kuala Selangor. The landfill started operations on 1st January 2007. The landfill is approximately 160 acres big and is designed with a capacity to hold 6 million tons of waste. Currently approximately 2000 tons of waste is disposed off at the landfill every day, and to-date 4.1 million tons of waste has been added to the landfill. Observed in the landfill site that there are several ponds containing leachate, raw untreated leachate and treated leachate. Leachate samples were collected from raw leachate pond andstored at temperature 4^oC to keep the wastewater characteristic unchanged.

Sample analysis

Samples collected were characterized and tested accordingly for few selected parameters: pH, color, chemical oxygen demand (COD), ammonium nitrogen (NH_3-N), Total phosphorus (Total-P), Total dissolved solid (TDS) and conductivity. Tests were conducted according to the standard methods for the examination of water and wastewater[13]. COD, color, NH_3N and total P (PO_4^{3-}) was measured using a HACH DR2400 spectrophotometer. TDS and conductivity were measured by using conductivity meter (Cond 610). The pH was measured by a pH meter (EUTECH pH 2700).

Preparation of electrodes

The composite electrodes at the composition of M85PVC15 were prepared accordingly, as already discussed by other author elsewhere, by mixing together a weighed portion of metals powder with PVC in 4 mL tetrahydrofuran (THF) solvent and swirled flatly to homogeneous followed by drying in an oven at 100°C for 3 hr. The mixture was then placed in 1 cm diameter stainless steel mould and pressed at 10 ton/cm[14]. A typical pellet contained approximately 85% (w/w) metals powder and 15% (w/w) of PVC polymer. The total weight of pellet obtained is approximately 1 g. The pellets were connected to silver wire with silver conducting paint prior covered with epoxy gum. The ratio of Pt, Al, Fe, Co, Cu, Ag and Ni powder with PVC in the prepared electrode are as summarized in Table 1.

Table 1: Ratio and composition metals powder and PVC for electrodes prepared

Electrode	Ratio metals powder: PVC	Metals powder (g)	PVC (g)
Pt_{85}-PVC_{15}	85:15	0.85	0.15
Ag_{85}-PVC_{15}	85:15	0.85	0.15
Cu_{85}-PVC_{15}	85:15	0.85	0.15
Ni_{85}-PVC_{15}	85:15	0.85	0.15
Al_{85}-PVC_{15}	85:15	0.85	0.15
Fe_{85}-PVC_{15}	85:15	0.85	0.15
$Co_{85}PVC_{15}$	85:15	0.85	0.15

The experimental setup

The electrochemical cell is consisted of a DC power supply (CP x 200 DUAL, 35 V 10A PSU) and a glass beaker (100 mL) completed with M_{85}-PVC_{15} composite electrode as an anode and stainless steel (10 mm × 10 mm) as cathode. The electrodes were placed parallel to each other in the electrolytic reactor, with the distance between the cathode and anode was approximately 1 cm. The stirrers was used in electrochemical cell to maintain an unchanged composition. Electrochemical cell as can be seen in Fig. 1.

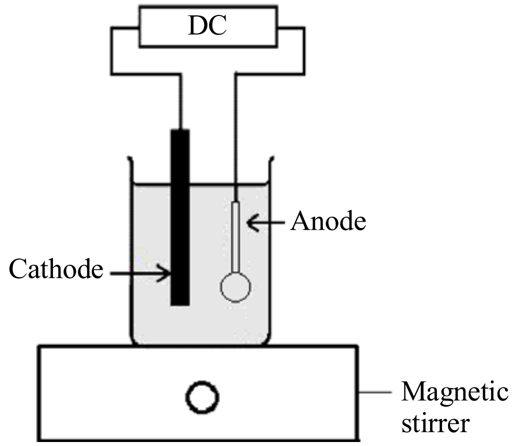

Fig. 1: Electrochemical cell setup

Experimental procedures

The experiment equipment was consisted of a DC power supply, glass reactor, and magnetic stirrer than added a known amount of solid NaCl as supporting electrolyte NaCl to 50 mL of leachate. During each experiment, mixing of the reactor contents was maintained to keep the solution homogenous using magnetic stirrer. All experiments were carried out at lab scale. The removal efficiency (%R) of landfill leachate can be obtained at any time, with respect to its initial values Eq. (2)[15]. The energy consumption (Esp) was calculated during process Eq. (1)[16].

$$Esp = \frac{E\,I\,t}{3600\,V} \qquad \ldots(1)$$

Where Esp, energy consumption in Wh/L, E is the applied voltage, I is the current intensity (A), t is the electrolysis time (s), V is the volume of the sample (L).

The removal percentage for COD, color and NH_3N was calculated according to Eq. (2).

$$R\,\% = \frac{A_o - A_t}{A_o} \times 100 \qquad \ldots(2)$$

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

RESULTS AND DISCUSSION

The characteristic of raw leachate

The characteristic of raw leachate was analyzed to determine the selected parameters pH, color, chemical oxygen demand (COD), ammonium nitrogen (NH_3-N), Total phosphorus (Total-P), Total dissolved solid (TDS), conductivity, biochemical oxygen demand (BOD), Total suspended solid (TSS) as shown in Table 2.

Table 2: Characterization of raw leachate samples collected from Jeram Sanitary Landfill

Parameter	Value
Color (Pt-Co)	14960
COD (mg/L)	49000
BOD_5 (mg/L)	14790
NH_3-N (mg/L)	3800
Total-P (mg/L)	200
pH	8.65
TDS (ppt)	28.09
TSS (mg/L)	2460
Conductivity (mS/cm)	29.67

Effect of electrode materials

Different electrode materials affect on the performance of the electrochemical oxidation process. In this study, several electrodes of metals powder consisting of platinum (Pt), aluminum (Al), iron (Fe), cobalt (Co), copper (Cu), silver (Ag) and nickel (Ni) with PVC as anode were fabricated. The best electrode base on color removal and electrochemical stability under the operating condition of applied voltage 10V, electrolysis time 90 min, NaCl concentration 1.2 % (w/v) was chosen compared with the metal plates[17]. The results obtained are summarized in Table 3.

Table 3: Electrochemical oxidation of landfill leachate using various composite electrodes and metal plates as an anode and stain steel as cathode

Electrode	Decoloring time (min)	Decoloring percentage (%)	Observation	
			Electrolysis product	Anode
Pt	120	56	Yellowish solution	Unchanged
Pt_{85}-PVC_{15}	90	99	Clear solution	Unchanged
Ag	120	9	Color unchanged	Slightly corroded
Ag_{85}-PVC_{15}	90	68	Yellowish solution	Slightly corroded
Cu	120	6	Color unchanged	Completely corroded
Cu_{85}-PVC_{15}	90	71	Yellowish solution	Slightly corroded
Ni	120	5	Color unchanged	Completely corroded
Ni_{85}-PVC_{15}	90	47	Yellowish solution	Completely corroded
Al_{85}-PVC_{15}	90	66	Yellowish solution	Unchanged
Fe_{85}-PVC_{15}	90	72	Yellowish solution	Slightly corroded
$Co_{85}PVC_{15}$	90	82	Clear solution	Unchanged

The best color removal (99%) was observed during 90 min. using Pt_{85}-PVC_{15} electrode compared with others. Meantime, the result of color removal usingPt_{85}-PVC_{15}, Al_{85}-PVC_{15}, Fe_{85}-PVC_{15}, Co_{85}-PVC_{15}, Cu_{85}-PVC_{15}, Ag_{85}-PVC_{15} and Ni_{85}-PVC_{15} electrodes were 99, 66, 72, 82, 71, 68 and 47%, respectively. The selection of the best electrode is not only to remove the color, but also on the basis of chemical stability. Because electrode used in any electrochemical process must be stable chemical and physical, as shown in Table 3[18]. Pt_{85}-PVC_{15} electrode was selected for further experiments.

Effect of supporting electrolyte concentration

The electrical conductivity of the solution is an important parameter for saving electric energy[19]. To increases the electric conductivity of the solution, NaCl was used as the supporting electrolyte due to its capability not only as a conductor but also able to enhance the degradation efficiency and shortened the reaction times due to the formation of hypochlorite ion, OCl^-.[20,21] It is apparent that increasing the Cl^- may increases the color removal due to the increased mass transport of chloride ions to the anode surface and also increased diffusion in the diffusion layer of the anode (Fig. 2). As a result, more amount of hypochlorite ion will be generated[22]. Thus, higher concentrations of hypochlorite ion were able to oxidize and degrade more organic molecules in landfill leachate[23]. A percentage of 1.2 % (w/v) of NaCl concentration was considered as an optimum electrolyte concentration due to highest color removal in landfill leachate (Fig. 2).

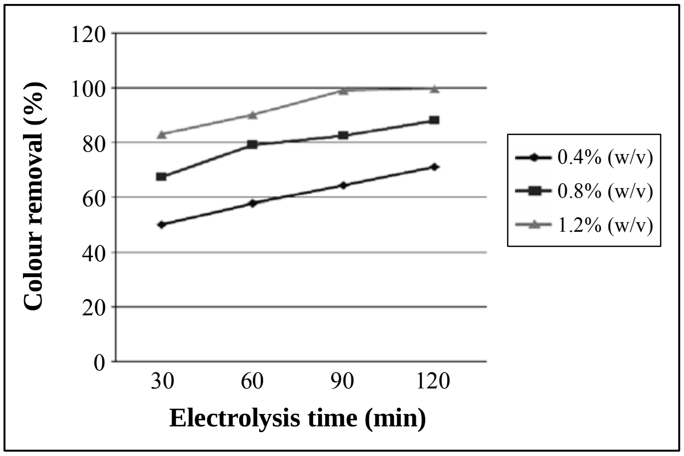

Fig. 2: Effect of NaCl concentration on color removal of landfill leachate

Effect of applied voltage

Electrolysis was conducted using different applied voltage values to investigate the effect of applied voltage on the color removal of landfill leachate. Fig. 3 shows that color removal rates for 4, 8, 10 and 14 V were determined to be 79, 84, 99 and 99.7%, respectively, for the 90 min treatment time.

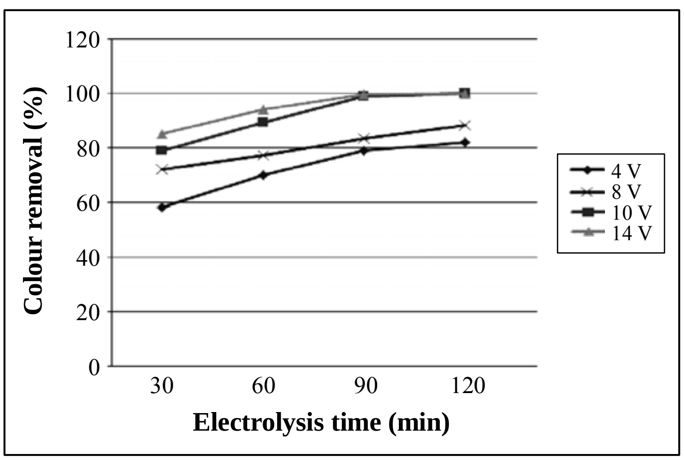

Fig. 3: Effect of applied voltage on color removal

This is due to production of oxidant, hypochlorite ion in the bulk solution. Increasing generation of oxidant is proportional to current density, which eventually increases the pollutant degradation[24,25]. The increase in hypochlorite ion approaches equilibrium with degradation of organics present in the effluent[26]. Increasing the applied voltage of the electrochemical cell followed by the production of more electron, which results in increasing the rate of overall reaction[27]. But more energy would be consumed at higher applied voltage. The results in Fig. 4 showed that 10 V as the optimal electrical potential for the electrochemical oxidation process. Because it ensures the removal rate accompanied with reducing the electrical energy to reach desired color removal, Therefore, 10 V of applied voltage has been selected for the further experiments.

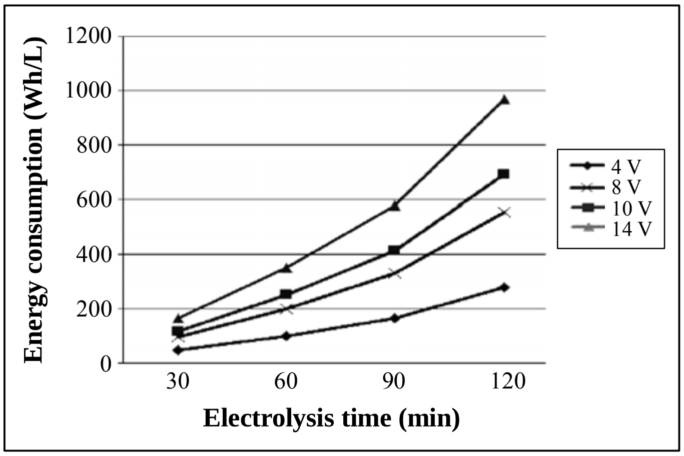

Fig. 4: Effect of voltage on consumption energy

Effect of electrolysis time

Effect of electrolysis time on color removal under the optimal conditions was investigated. As shown in Fig. 5, the color removal percentage rapidly increased with the increased of electrolysis time upto 90 mins. After 90 mins of electrolysis time, color removal percentage was slowly increased until upto 99%. The color removal efficiency depends directly on the concentration of electrochemical generated of hypochlorite ion in the bulk solution. When the electrolysis time was longer, more hypochlorite ion will be produced in solution under fixed current density[28]. Therefore, color and COD value in the solution were reduced in higher concentration of hypochlorite.

Treatment of raw leachate using electrochemical methods at optimum conditions

Base on optimization studies to the treatment of landfill leachate, the optimum operating conditions are; 10 V, raw pH, NaCl with concentration 1.2% (w/v), Pt_{85}-PVC_{15} electrode and 90 min electrolysis time (Table 4).

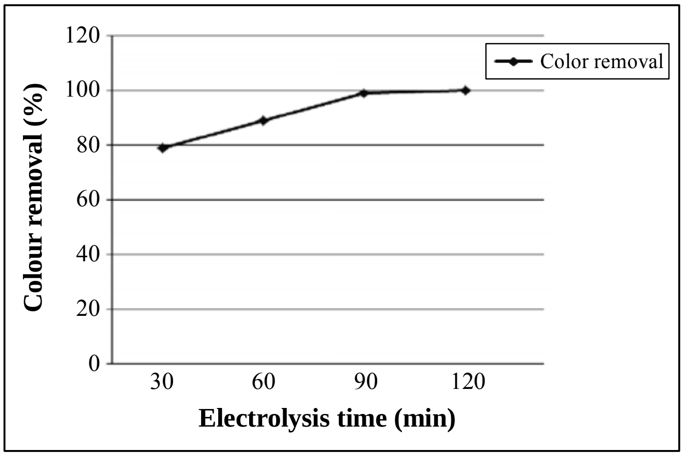

Fig. 5: Effect of electrolysis time on color removal

Table 4: Optimum operating condition for landfill leachate treatment using Pt_{85}-PVC_{15} electrode

Electrodes working (anode)	Pt_{85}-PVC_{15}
Counter (cathode)	Stainless steel
Supporting electrolyte	1.2 % (w/v) NaCl concentration
Applied voltage	10 V
Electrolysis time	90 mins

The results obtained show that the electrochemical oxidation process is able to reduce the color, COD and NH_3N values in landfill leachate. From those three parameters under studied (color, COD and NH_3N), the removal percentage were not less than 99, 98 and 91%, respectively (Fig. 6).

The reduction in these parameters value was due to the breaking of large molecule to a small molecule, which is easier to be oxidized chemically or biologically by electrochemical oxidation technique[29]. This breaking process was supported by the existence of self-generated hypochlorite ion, which is able to reduce the concentration of organic compound available[30].

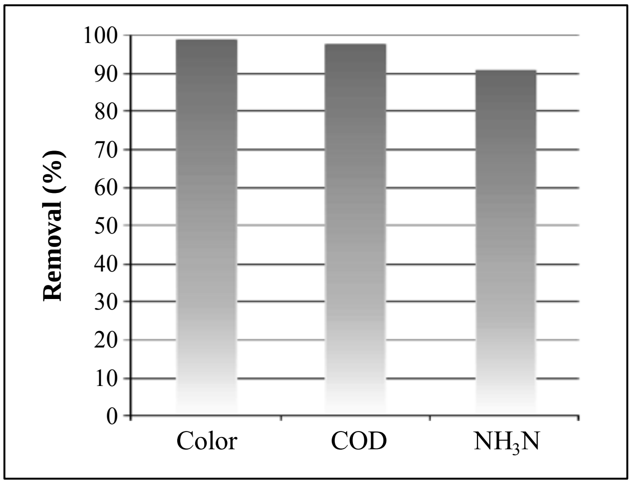

Fig. 6: The percentage of removal

The electrochemical oxidation mechanism for COD removal

The following equations represent the reactions that occurs in the electrochemical oxidation of landfill leachate.

The anode: $2\ Cl^- \longrightarrow Cl_2 + 2\ e^-$

$4\ OH^- \longrightarrow O_2 + H_2O + 4\ e^-$

The cathode: $2\ H_2O + 2\ e^- \longrightarrow H_2 + 2\ OH^-$

In the bulk solution:

$Cl_2 + H_2O \longrightarrow H^+ + Cl^- + HOCl$

$HOCl \longrightarrow H^+ + OCl^-$

Leachate + $OCl^- \longrightarrow CO_2 + H_2O + Cl^-$ + Product

The electrochemical oxidation mechanism for NH_3N removal

High NH_3N removal efficiencies can be achieved by elecrooxidation process. Removal of nitrogen compounds can be explained in mechanism by using this process.

$2\ Cl^- \longrightarrow Cl_2 + 2\ e^-$

$Cl_2 + H_2O \longrightarrow HOCl + H^+ + Cl^-$

$2\ NH_3 + 3\ HOCl \longrightarrow N_2 + 3\ H^+ + 3\ Cl^- + 3\ H_2O$

The electrochemical oxidation for color removal

As color is not a pollutant parameter alone, that is known that it is an important pollutant indicator. For this purpose, also color removal is investigated beside organic and nitrogen compound pollutants. Together with the electro oxidation process, also strong oxidation process occurs so that color causing pollutants can be removed. Findings that are achieved in this study are satisfactory for a highly colored landfill leachate.

CONCLUSION

Based on the experiment of treatment of leachate by electrochemical oxidation, this paper studies the factors affecting the removal efficiencies. The results indicate that electrochemical oxidation process can be used to the leachate preprocessing. Under conditions of Pt 85 PVC 15 electrode, 10V applied voltage, 1.2% (w/v) Cl^- concentration, 90 min electrolysis time and unchanged the raw pH, the removal efficiencies of color, COD and NH_3N 99, 98 and 91%, respectively.

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