

Synthesis and Electrochemical Treatment of Leachate in Malaysia

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Abstract: Electrochemical techniques have been applied extensively to treat the landfill leachate. Electrocoagulation (EC) and electrochemical oxidation (EO) are the treatments that are being concerned in this research. The goal of this research is to study the efficiency of pollutants removal (BOD₅, COD, TSS, NH₃-N, and colour) of landfill leachate by applying EC and EO. Based on the result, longer EC time would result in a higher pollutant removal efficiency. In the sludge's settling characteristic determination, the longer the EC duration, the SVI increases and the settling velocity of the sludge decrease. On the other hand, the EO was carried out in electrochemical cells using a graphite carbon as anode and cathode. Results have shown that by increasing the addition of Cl⁻ and the current density, the removal percentage of pollutants increases. For pH, acidic condition favours the removal of COD and colour, while alkaline condition favours the removal of BOD₅, NH₃-N and TSS. Apart from EC and EO treatment, the research has deduced that 43.61% and 46.18% of COD and BOD₅ respectively exists as particulate matter, and they can be removed as solids through filtration. Moreover, by treating the particulate-free leachate with EC proved that co-reaction occurs during EC treatment.

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1. Introduction

Growth in the population and new consumption method have been leading to the production of huge amount of municipal solid wastes. After these wastes are being produced, they usually send to sanitary landfills, as this consume relatively low cost, and the procedure is simpler [1]. However, when the rainwater percolation through the wastes, the water extracts several pollutant materials. Finally, a very complex sanitary landfill leachate has produced [2].

Leachate generated from municipal landfills often contain organic and inorganic compounds, heavy metals, ammonium and many more soluble and insoluble

compounds [3]. Landfill leachate usually contains high value of chemical oxygen demand (COD), biochemical oxygen demand (BOD), colour, total suspended solid (TSS) and ammonia nitrogen (NH₃-N). The composition of the landfill leachate varies due to landfill characteristics, such as climate, landfill cover, type of waste received, landfill environment and many more [2]. Due to its complexity, landfill leachate can be considered as one of the major environmental problems concerning water pollution. Proper leachate management and treatment should be carried out in order to prevent water resource contamination, especially underground water and water in the soil. Conventional treatments such as

biological, chemical and physical methods are ineffective and expensive to treat leachate [4].

There are several advantages of electrochemical treatment. It is characterized by simple equipment, easy operation and brief retention time. Besides, the process has high robustness as it can be terminated at any time by cutting the power supply. Moreover, electrochemical treatment does not need high temperature and high pressure. As consequences, volatilization and discharge of unreacted wastes are avoided. Next, it is environmental compatibility as it does not require any addition of artificial chemical. Besides, it exhibits its versatility as it can deal with many pollutants and treat quantities from microliters to millions of litres [5]. Electrocoagulation (EC) and electrochemical oxidation (EO) are the two electrochemical treatments that are being concerned in this research.

EC treatment of wastewater has been used since the 20th century. Yet, it was still with limited success and popularity back then. In the last decade, South America and Europe had been increasingly using this EC technology for industrial wastewater treatment. In North America also, EC was used for wastewater treatment in pulp and paper industries. Nowadays, EC has been used in many fields including treatment of water containing dyes, oil wastes, chemical and mechanical polishing waste, foodstuff waste, and also organic waste from landfill leachates [6]. EC is one of the simple and efficient electrochemical methods for water and wastewater treatment. An EC reactor is build-up with an electrolytic cell with two electrodes [7]. One is the anode and the other is the cathode. The electrode is made up of conductive metal plates, such as aluminium and iron. When the electricity is passed through the electrodes, the coagulant is produced *in situ* by electrolytic oxidation of the anode material, at an appropriate pH, into insoluble metal hydroxide that has the potential to remove a variety of pollutants [8]. The metal hydroxide species produced have the ability to neutralize the electrostatic charges on suspended solids, particulate matters, and oil droplets to facilitate coagulation or agglomeration. Subsequently, the coagulated particles are being separated from the liquid phase by either sedimentation or electroflootation [9].

On the other hand, electrochemical oxidation (EO) has been showing as a good alternative in electrochemical treatment. EO treatment is very popular and has received a great attention in wastewater treatment and organic pollutants oxidation in recent years. EO is widely applied in a variety of treatments, including treatment of textile wastewater, coke plant wastewater, tannery wastewater and much more [10]. This method also applied for the treatment of landfill leachate [11]. In EO, pollutants are being degraded by two ways, either direct oxidation or indirect oxidation [12]. For direct oxidation, pollutants are absorbed on the anode surface, being oxidized and then being destroyed by the anodic transfer reaction. On the other hand, for indirect oxidation reaction, strong oxidants are being

generated *in situ* and utilized immediately. For example, hypochlorite/chlorine, hydrogen peroxide and ozone are being electrochemically generated. These strong oxidants are responsible for destroying the pollutants in the wastewater or leachate by oxidation reaction [13]. The set-up for EO is almost same as EC. The only difference between them in the setting up is the type of electrode. Anode is the electrode that is being concerned in EO, as oxidation will take place at the anode. Anode can be differentiated into two types, one is “active” and one is “non-active”. Some of the examples of “active” anodes are Pt, IrO₂ and RuO₂. On the other hand, examples of the “non-active” anodes are PbO₂, SnO₂ and boron-doped diamond (BDD). The reason the anode is called “non-active” electrode is due to it does not provide any catalytic active site for the adsorption of reactants and/or products from the aqueous medium [14].

Although EC and EO have been studied widely, more research should be done to further understand and optimise these treatments. Electrocoagulation is known as a method which removes the pollutants by neutralising the particulate/colloidal matter. Yet, many studies did not measure to what extent the fraction of pollutants (BOD₅, COD, TSS, NH₃-N, colour) are contributed by the particulate/colloidal matters and how much are contributed by the soluble matters in the leachate. Moreover, the removal of certain soluble pollutants (colour and NH₃-N) by EC is still a question to be answered. More researches have to be done on EC treatment to test whether it only can remove particulate/colloidal matters, or there are some co-reactions happens that leads to removal of soluble pollutants.

Besides, most of the studies of EC today had focused on finding the best operating conditions in removing the pollutants, such as the optimum time, initial pH, current density, electrode material, etc. Yet, the condition, characteristic and settling time of the sludge does not get a lot of attention in the studies. Ricordel and Djelal [15] had a contrary statement in their research. They stated that with longer time of treatment, higher pollutant removal efficiency was obtained. Again, they found that with a short EC time (30 min), larger and denser flocs with good settling time had been produced. Therefore, the relationship between the removal efficiency and the characteristic of sludge have not been studied enough. In order to make the reactor design practical, the pollutant removal efficiency and the sludge characteristic are equally important. The best operating time to produce high removal efficiency with acceptable sludge settling should be determined. In EO, high-quality anode material, Boron-Doped Diamond (BDD), TiRuSnO₂, PbO₂/Ti, etc. seem to have good efficiency in EO treatment process, but these electrodes are quite rare and expensive. These electrodes may not be that suitable to apply in a large treatment plant. A cheaper and highly available alternative, such as graphite anodes, needs to be taken into account. Although graphite electrode has lower efficiency compared to the others, it may give

promising result if the operating condition is well tuned. Therefore, studies have to be done to determine whether graphite anodes are suitable as alternatives to other anodes. By understanding the fundamental mechanism and theory behind the electrochemical treatment, its advantages can be maximized and provide a better leachate treatment process.

2. Materials and Methods

2.1 Materials

Materials needed for this research is the sample of leachate, hydrochloric acid, sodium hydroxide and salt (NaCl). The leachate was sampled from Seelong Sanitary Landfill, Johor, Malaysia during normal day condition. This sanitary landfill started its operation in the year 2003. The collected samples were stored at a temperature of 4 °C in order to minimise biological and chemical reactions. The leachate temperature was increased to room temperature before proceeding with experimental purposes. The NaCl salt was used to increase the chloride concentration for EO leachate treatment processes. Hydrochloric acid and sodium hydroxide were used to adjust the pH of the sample leachate for experimental purposes. Moreover, the electrodes were washed by using hydrochloric acid to remove impurities before the experiment started.

2.2 Experimental Rigs

In this research consist of two experimental rigs, which is electrochemical cell and filtration system. EC and EO equipment set up were carried in batch mode. Leachate was placed in a beaker. The electrodes were connected to positive and negative pole of the DC power supply and dipped inside the leachate. Different electrodes were used for EC and EO. Electrode for EC would be aluminium and EO would be graphite carbon. Magnetic stirrer, which has the speed of 100 rpm, was placed in the jar to provide slow mixing. In order to wash away the impurities, the electrodes were washed with dilute hydrochloric acid before the conduction of experiment. The electrochemical process was carried out at room temperature. The leachate was filtered using filter paper in order to remove all the particulate matters. Comparison of results is made before and after the filtration to study the contribution of BOD, COD, ammonia nitrogen, and colour by the particulate matters in leachate.

2.3 Experimental Analysis

Different pollutants were tested with different method. The BOD was measured by conducting BOD₅ test. COD was tested by HACH Method 8000. The TSS was measured through laboratory test by removing moisture content from the sample. Lastly, NH₃-N was tested by HACH Method 10031. Besides that, sludge settling test were also conducted in this study. After an EC run, leachate was further submitted to settling tests to identify sedimentation sludge settling phases. 1 L of

treated leachate was left in the glass jar where the EC take place. The height of the liquid/sludge interface was recorded at regular intervals and sludge settling was left for 1 hour. Sludge volume index (SVI) and settling velocity had been determined from settling test.

2.4 Experimental Procedure

2.4.1 Determination the fraction of pollutants contributed by particulate matters (Phase 1)

The leachate sample was filtered by filter paper to remove the particulate matters. The compositions of leachate were compared between the readings before filtration and after filtration. The fraction of pollutants contributed by the particulate matters in leachate were determined.

2.4.2 Determination of co-reaction in EC (Phase 2)

Particulate-free leachate was used for co-reaction determination. Electrochemical cell was being set up. Aluminium was used as the electrode for EC. The volume of leachate is 200 mL and the current density of the experiment is 20 mA/cm². The EC process was carried out for 30 minutes.

2.4.3 Determination of optimum time duration in EC (Phase 3)

Electrochemical cell was being set up. The volume of leachate was 1 L. Aluminium was used as the electrode for EC. pH 7 was used as the initial pH and current density of 60 mA/cm². The EC processes were running separately for 30, 60, 90, 120 minutes. After the EC, the height of the liquid/sludge interface was recorded at regular intervals and sludge settling was left for 1 hour. SVI and settling velocity were determined from settling test.

2.4.4 Determination of optimum operating conditions in EO by using Graphite Electrode (Phase 4)

Graphite carbon was used as the electrode for EC. Time duration for EO was fixed at 180 minutes. Different current density, pH and chloride ion concentration were tested for the optimum operating condition. Current density was tested for 20, 40, and 60 mA/cm² to determine the removal efficiency. The addition of Cl⁻ was fixed at 8000 mg/L and pH of 7. Cl⁻ concentration were added for 2000, 5000, 8000, and 11000 mg/L to determine the removal efficiency. Current density was fixed at 60 mA/cm² and pH at 7. Initial pH was tested for 5, 6, 7 and 8 to determine the removal efficiency. Current density was fixed at 60 mA/cm² and Cl⁻ addition at 8000 mg/L.

3. Results and Discussions

3.1 Leachate Composition

The leachates were collected from Seelong Sanitary Landfill during normal days. Table 1 depicts the

composition of leachate collected from Seelong Sanitary Landfill.

Table 1 - The Concentrations of BOD, COD, Ammonia Nitrogen, TSS and pH of Leachate Obtained from Seelong Sanitary Landfill (SSL)

No.	Parameters	Value
1.	pH	8.36
2.	COD (mg/L)	21900
3.	BOD ₅ at 20°C (mg/L)	825
4.	BOD ₅ /COD	0.04
4.	TSS (mg/L)	1700
5.	Ammoniacal-Nitrogen (mg/L)	2735
6.	Colour	Dark Black

3.2 Fraction of Pollutants caused by Particulate Matters in Leachate

Table 2 depicts the treated parameters after filtration. Filter paper was used for the filtration experiment to determine the contribution of BOD, COD, ammonia nitrogen, and colour by the particulate matters in leachate.

Table 2 - Treated Parameters by Filtration

Parameters	After Treatment	Removal Efficiency (%)
COD (mg/L)	12350	43.61
BOD ₅ (mg/L)	444	46.18
Ammoniacal-Nitrogen (mg/L)	2500	8.59
Colour	Dark Black	-

The COD and BOD₅ in the filtered solution were tested to determine the efficiency of filtration in removing COD and BOD₅ in the form of suspended solids. The percentage of removal in COD and BOD₅ are 43.61% and 46.18% respectively. It can be deduced that about 45% of COD and BOD₅ exists as particulate matter, and they can be removed as solids through filtration. Regarding the ammonia nitrogen, although it usually exists in soluble form, ammonia nitrogen may still have the probability to be presented in insoluble form as ammonium salt complexes [16]. This can be explained by the 8.59% removal of ammonia nitrogen by filtration. For colour, humic acid can be considered almost soluble [17]. Therefore, from the filtration, no visible colour changed has been observed.

3.3 Co-reaction in EC

Particulate-free leachate will be used for co-reaction determination. The filtered leachate after the filtration experiment was used in this experiment. Table 3 depicts the treated parameters of filtered/particulate-free

leachate by EC. From the results, it has proven that co-reaction indeed take place during the EC, due to the removal of pollutants in particulate-free leachate.

Table 3 - Treated Parameters of Particulate-Free Leachate by EC

Parameters	Before Treatment	After Treatment	Removal Efficiency (%)
COD (mg/L)	12350	10450	15.36
BOD ₅ (mg/L)	444	373	16.00
NH ₃ -N (mg/L)	2500	2230	10.80
Colour	Dark Black	Brown	

3.4 Optimum Reaction Time Determination in EC

Fig. 1 illustrated the findings on the effect of reaction time towards the removal efficiency of BOD, COD, TSS and NH₃-N. and Fig. 2 portrayed the effect of reaction time on the removal efficiency of colour. From Left to Right: Raw Leachate, 30 mins, 60 mins, 90 mins, and 120 mins.

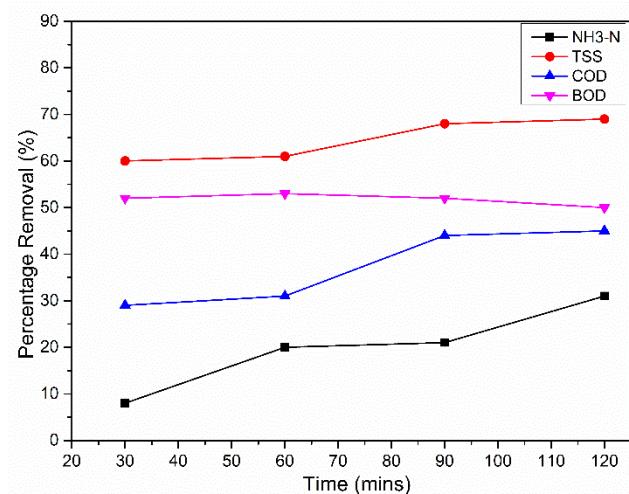


Fig. 1 - The effect of reaction time on the removal efficiency of BOD, COD, TSS and NH₃-N



Fig. 2 - The effect of reaction time on the colour. From left to right: Raw leachate, 30, 60, 90, and 120 mins, respectively.

Longer EC time would result in a higher pollutant removal efficiency. Longer EC duration led to higher coagulant concentration, which removes more pollutant from the leachate. Based on Fig.1, for COD, BOD₅ and TSS, the time to reach the constant removal percentage is 90 mins, but the removal percentage NH₃-N still increases after 90 mins. The increase in the removal percentage of NH₃-N after 90 mins can be explained by EO co-reaction that occurs during EC. Different researcher had shown different result in for the EC to reach the constant removal efficiency. Li and co-workers [18] stated that an increase in the time from 5 to 90 minutes yield an increase in the efficiency of COD removal from 18.8 to 45.0% and the NH₃-N removal from 9.4 to 44.1%, but the removal does not change significantly after 90 min. On the other hand, Ricordel and Djelal [15] results show that it needs about 120 mins to 150 mins to reach the constant removal efficiency. Overall, it can be concluded that the optimum reaction time lies within 90 mins to 150 mins. More research should be done on the composition of the leachate so that an optimum reaction time can be estimated.

3.5 Sludge Characteristic in EC

Sludge settleability can be expressed by sludge volume index (SVI) and settling velocity. The expression of SVI is shown below:

$$SVI \left(\frac{mL}{g} \right) = \frac{H_{30}}{H_0 \times SS} \times 1000 \quad (1)$$

where H₃₀ is the sludge height after 30 min settling (cm), H₀ is the initial height of the electrocoagulated waste in the settling column (cm) and SS is the initial sludge concentration after EC treatment in g/L.

Besides, the sludge settling velocity is an important parameter for identifiable visible solid/liquid interface [15]. The expression is shown in Equation (2).

$$Settling\ velocities = \frac{\Delta V}{\Delta t} \quad (2)$$

where ΔV is change in volume, Δt is the change in time.

Table 4 - Parameters of SVI and settling velocity at different EC duration

Parameters	Time (mins)			
	30	60	90	120
SVI (mL/g)	39.68	101.32	166.11	201.21
Settling Velocity (L/mins)	0.00576	0.00547	0.00518	0.00518

From the result obtained, it has displayed that the SVI increase from 39.68 mL/g to 201.21 mL/g with the increase in the EC reaction duration from 30 mins to 120 mins. This result has met an agreement with the result found by Zodi and co-workers [19]. They reported that SVI values increased with the treatment period with Al electrodes, which increased from 124 to 267 mL/g as the

EC period varies from 40 to 120 min. The increase in SVI may due to the increase in the coagulant concentration which directly lead to larger flocs size. Large flocs are the reason behind the difficulty in sedimentation and compaction of sludge.

3.6 Electrochemical Oxidation

3.6.1 The Effect of the Chloride Addition

Fig. 3 shows the effect of chloride addition on the removal efficiency of BOD, COD, TSS and NH₃-N. Meanwhile, Fig. 4 shows the effect of chloride addition on the colour.

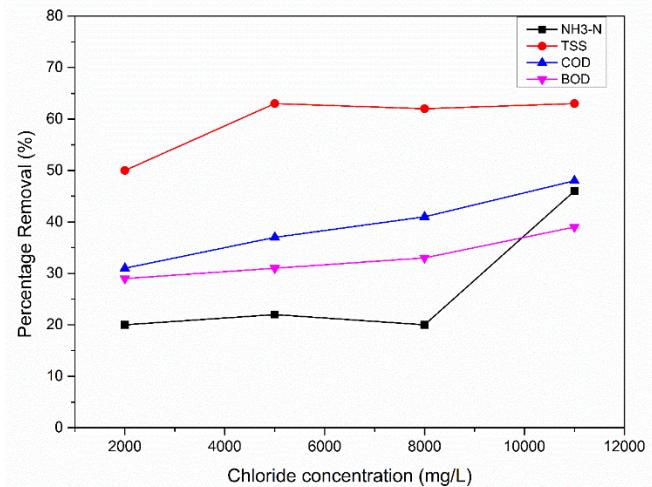


Fig. 3 - The effect of chloride addition on the removal efficiency of NH₃-N, TSS, COD and BOD.



Fig. 4 - The effect of chloride addition on the removal efficiency of colour. From left to right: raw leachate, 2000, 5000, 6000 and 11000 mg/L, respectively.

The result has shown that increase in the Cl⁻ addition would increase the removal efficiency of COD, BOD, TSS and especially, NH₃-N, which had shown a drastic increase in the removal percentage. This result has shown similarity with a lot of literature being reported. The addition of extra Cl⁻ generally improves electro-oxidation of landfill leachate, which is explained by the enhanced indirect oxidation through higher chlorine/hypochlorite production [3],[20],[21]. Addition of chloride ion would result in a lower voltage applied through the electrode. Therefore, using high initial chloride concentrations can reduce the energy consumption, mainly due to an increase in the conductivity.

3.6.2 The Effect of the pH

Fig. 5 illustrated the effect of pH on the removal efficiency of BOD, COD, TSS and NH₃-N. Meanwhile, Fig. 6 shows the effect of pH on the colour reduction.

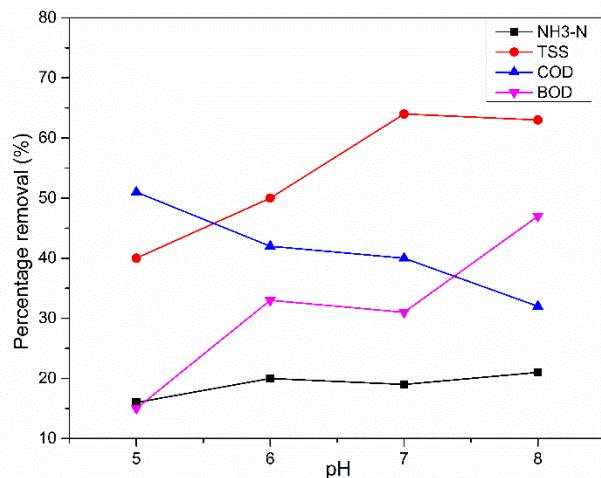


Fig. 5 - The effect of pH on the removal efficiency of NH₃-N, TSS, COD and BOD.



Fig. 6 - The effect of pH on the colour removal. From left to right: raw leachate, pH 8, 7, 6 and 5, respectively.

Based on the graph, two conclusion can be made. By increasing the pH from 5 to 8, the removal efficiency of BOD, TSS and NH₃-N increases while the COD and colour removal had shown a different trend. The situation above can be explained by different mechanism that occur during EO treatment at high pH and low pH. Acidic conditions significantly decrease the concentration of CO₃²⁻ and HCO₃⁻, which are the scavengers of •OH generated on anodes, and so enhances direct oxidation, while alkaline condition boosts the Cl⁻→Cl²→ClO⁻→Cl⁻ redox circulation to enhance the indirect oxidation [11]. The result of higher COD removal at low pH has been proven by the research from Vlyssides and co-workers [22]. They reported that a low pH favoured COD removal and energy consumption. Other researchers had also found pH 4.0 achieved at least 20% higher COD removal than pH 8.0 at an EO treatment [23]. These statements have proven that direct oxidation can remove COD more efficiently.

For colour, which causes by the humic acid presents in the leachate, theoretically can be oxidized by the

hydroxyl radical, •OH. Hydroxyl radical is known as the bleaching agent. Reaction of •OH with humic acids results in the release of low molecular weight acids, amino acids, ammonia and dissolved inorganic carbon (DIC) [24],[25]. As mentioned above, acidic conditions significantly decrease the concentration of CO₃²⁻ and HCO₃⁻, which are the scavengers of •OH generated on anodes, and so enhances direct oxidation. On the other words, at low pH, there are more •OH in order to remove the humic acid in the leachate, and subsequently, reduce the colour of the treated leachate.

At alkaline condition, it boosts the Cl⁻→Cl²→ClO⁻→Cl⁻ redox circulation to enhance the indirect oxidation, which means it would have higher concentration of chlorine/hypochlorite. Granum and Magnusson [26] reported that for the microorganism, the effect of hypochlorite increased by a factor of about 2.5 from pH 5 to 8, and by a factor of 6 from pH 8 to pH 9. This means that hypochlorite act as a better disinfectant in higher pH, and this statement has matched with the result shown in Fig. 5, which higher BOD removal in higher pH. On the other hand, Ammonia Nitrogen is effectively removed by indirect oxidation through chlorine and hypochlorite. According to Pérez *et al.*, [20] ammonium degradation takes place mainly due to the indirect oxidation through chlorine/hypochlorite.

3.6.3 The Effect of the Current Density

Fig. 7 demonstrated the effect of current density on the removal Efficiency of BOD, COD, TSS and NH₃-N and Fig. 8 shows the effect of current density on the removal efficiency of colour.

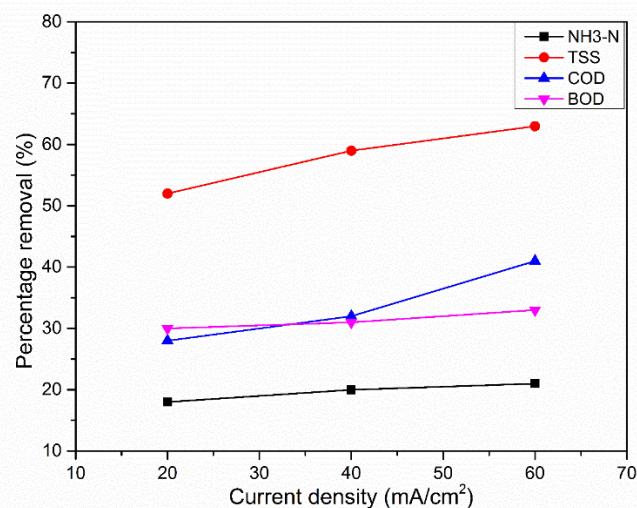


Fig. 7 - The effect of current density on the removal efficiency of NH₃-N, TSS, COD and BOD

Based on the result shown above, the percentage of pollutants removal increase with the increase of current density. It has been explained that by increasing the current density, the production of oxidizing species (chlorine/hypochlorite) would be increased as well [27].

From the result obtained, the highest percentage of removal by using carbon electrode is 41.32% in COD,

33.45% in BOD, 64.71% in TSS and 20.66% in ammonia nitrogen. Although increasing the current density may increase the pollutant reduction from leachate, the performance of carbon electrode is still incomparable with other electrode, such as the boron-doped diamond (BDD). By using BDD, samples were analysed for COD as control of the treatment method being its removal percentage nearly 90% after 180 min [28]. In the paper of Panizza and Martinez-Huitle [29], after 8 hours of experiment, results shows that BDD enables complete COD, colour and ammonium removal due to the electrogeneration of hydroxyl radicals from water discharge and active chlorine from chloride ions oxidation.



Fig. 8 - The effect of current density on the colour reduction. From left to right: raw leachate, current density of 20, 40 and 60 mA/cm², respectively.

4. Conclusion

In this study, electrocoagulation (EC) and electrochemical oxidation (EO) are the treatments that are being concerned. Longer EC time would result a higher pollutant removal efficiency. In the sludge's settling characteristic determination, the longer the EC duration, the SVI increases and the settling velocity of the sludge decrease. For EO, results have shown that by increasing the addition of Cl⁻ and the current density, the removal percentage of pollutants increases. For pH, acidic condition favours the removal of COD and colour, while alkaline condition favours the removal of BOD₅, NH₃-N and TSS. The treatment of landfill leachate using the combined system is a proper way to treat landfill leachate to produce clean discharge. This innovative treatment system is expected to reduce the water pollution caused by landfill leachate. This system is believed to bring benefits to human beings, including mitigating the problem of water pollution effectively.

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