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Removal of Suspended Solid, Colour and Ammoniacal-Nitrogen from Leachate Using Aerated Electrochemical Coagulation (AEC) Under the Influence Factors of pH and Electrolysis Duration

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Abstract: The leachate produced by the decomposition of waste in landfills contains toxic substances that pollute the environment and cause health concerns which proper leachate treatment is essential before leachate is directly discharge into the environment. The most important factors to be considered when adopting a leachate treatment are effluent removal performance and the ability of treated effluent to satisfy the standard limit discharge. This study investigates the optimum condition of Aerated Electrochemical Coagulation (AEC), a mix of physical and chemical treatments in removing suspended solid, colour and ammoniacal-nitrogen from leachate under the influence factors of pH and electrolysis duration. Stabilized leachate from Simpang Renggam Landfill was used as samples for this study. From the data analysis, the highest removal effectiveness was achieved at 10 minutes of electrolysis duration and pH 5 for removal of suspended solid, colour and ammonia-nitrogen with 93%, 93%, and 40%, respectively. Based on the data, integration of the aeration method into electrochemical coagulation in leachate treatment has shown enhancement of the removal efficiency of suspended solid, colour and ammoniacal-nitrogen. However, only suspended solid satisfy the standard leachate discharge.

Keywords: Aerated Electrochemical Coagulation (AEC), leachate, landfill, Johor

1. Introduction

Solid waste management issues, whether in small or large cities, are very challenging for authorities in a developing country as the systems are still adopting the conventional method. This is mostly due to the rising production of such solid waste and the burden placed on municipal budgets [1] as well as poor understanding of various factors that affect the entire handling system. Landfills are the most common method of disposing municipal and non-

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hazardous industrial solid waste mainly due to their simple operational and economic factor [2]. However, open and closed landfills harm the environment through leachate formation if improperly managed [3]. Leachate contains dangerous pollutants such as ammonia, nitrogen, heavy metals, organic matter and toxic compounds that will pass through soil and penetrate the surface water or groundwater with aid from the excessive rains, which lead to various environmental pollution cause health issues [4], [5]. Landfill activity associated with various environmental degradation that reduces the quality of our water resources and affecting life [6]. Therefore, leachate must be collected and appropriately treated before being discharged, as directly discharging leachate into the environment would cause severe problems for an extended period and irreversible harm to the environment. Furthermore, changes of leachate characteristic throughout its ageing process differentiate its from others wastewater.

Landfill leachates have three key groups such as young leachate, intermediate leachate, and stabilized old leachate. The biological process is effective for young leachates, which specifically to be less than five years old and is incompetent for stabilized old leachate [7]. Contrarily, physical and chemical treatments are favored for the treatment of stabilized old leachate and are not recommended for young ones [8]. The efficiency of various methods for leachate treatment depends on the leachate characteristics, which may hinder the application in certain cases. As for stabilized old leachate, the most effective treatments are chemical adsorption, reverse osmosis, and coagulation-flocculation [9]. However, among all treatment methods suitable for stabilized old elachate, coagulation is the simplest and economical method [10]. Nowadays, many treatment methods for leachate have recently arisen due to new and improved technologies such as biological, chemical, physicochemical, or a combination of treatments [11]. Furthermore, the chosen treatment should be adaptable enough to stay useful if regulations, leachate properties, and economic situations change. According to Kurniawan et al. [12], the essential considerations in choosing a leachate treatment are its effluent removal performance and treatment cost and whether the treated effluent can meet the effluent limit at a reasonable cost. Due to the high concentrations of contaminants in intermediate and stabilized old landfill leachate and its low biodegradability, a combination of physical and chemical treatments is strongly recommended for leachate treatment, which can enhance removal efficiencies and minimize energy consumption [13]. Different types of leachate treatments have their own unique and signature agents to work efficiently for each category of leachate that is affected by the decisive factors of landfill age, biodegradable ratio, and various parameters. It was observed that, when the treatment techniques are employed in standalone mode, they are unable to meet the acceptable water quality criteria, nevertheless when the treatment techniques are used in combination, then it meets the stringent criteria for disposal [14]. The integrated biological and physicochemical treatment methods are viewed as critical for adhering to sustainable release limits and ensuring environmental protection by achieving satisfactory pollutant removal efficiency. However, several established methods have high capital operational costs and complex operations [15]. Therefore, much effort has been made for further development of simple and economical treatments.

Electrocoagulation (EC) as a leachate treatment has gotten much attention in recent decades. EC treatment has evolved as an environmentally benign method that produces less sludge, requires no additional chemical additives, and has a minimal footprint without affecting treatment efficiency [16]. Meanwhile, AEC is integration of air into the EC system. Air is introduced into the system to increase the amount of oxygen available for the reaction. The aeration technique was conducted by supplying additional oxygen to the leachate sample. Aeration increases the amount of dissolved oxygen in water, reduces the number of gaseous components in waste, and acts as an oxidizer, allowing easily oxidized chemicals to have enough oxygen to undergo oxidation reactions and oxidized substances to separate from the solution [17]. Kumar et al. [18] conducted electrocoagulation tests on composite waste with and without the addition of oxygen, discovering that adding extra oxygen resulted in higher colour and COD removal. Before treatment, the pH of the solutions is considered a critical parameter because it affects solution conductivity, electrode dissolution, hydroxide speciation, and colloidal species potential [19]. Even though the EC process appeared to function successfully throughout a wide pH range in most studies, there was a relatively narrow pH range where the process performed effectively, which was typically found to be near neutral pH values [20]. Another critical parameter that influences the EC treatment is electrolysis duration. In a review of treatment for textile wastewater reported by Naje et al. [19], the efficacy of pollution removal improves as the electrolysis duration is extended. After reaching the optimum electrolysis time, the removal rate remains constant. Thus, the main objective of this study is to investigate the optimum condition of AEC in removing suspended solid, colour and ammoniacal-nitrogen from stabilized leachate under the influence of pH and electrolysis duration. Then, determine the efficiency of AEC by comparing the treated effluent with the standard leachate discharge according to Malaysia Environmental Quality Act (EQA) 2009 [21].

2. Materials and Methods

2.1 Study Area

Raw leachate samples were collected from the Simpang Renggam Landfill (SRL) site in Simpang Renggam, Johor. It had been operated for over ten years with a land area of 28 hectares near an oil palm plantation before being permanently closed for solid waste disposal from three main districts in Johor, namely Batu Pahat, Kluang, and Simpang Renggam. A landfill will continue to produce leachate even after it has been closed for several years, which requires ongoing monitoring and maintenance to avoid any additional environmental contamination that could arise after the physical closure. Fig. 1 shows the location of Simpang Renggam Landfill site for this study.

2.2 Leachate Sampling

A leachate sample was taken from Simpang Renggam Landfill. Leachate was immediately transported to Universiti Tun Hussein Onn Malaysia Micro Pollutant Research Centre (MPRC) laboratory and stored in a chiller at a temperature of 4°C before analysis. The sampling and storage of leachate samples were performed according to Standard Method for Examination of Water and Wastewater 2008. The sample collected was used for leachate characterization and optimization of pH and electrolysis duration to obtain the optimum condition of AEC.

2.3 Equipment and Reagent

Table 1 shows the equipment, reagent and method used in analysis of parameter suspended solid, turbidity, pH, colour, ammonical-nitrogen, DO, BOD, COD and AEC treatment.

Table 1 - Equipment and reagent used in this experiment

Parameter	Equipment	Reagent	Standard Method
Suspended solid	DR6000		APHA 2540D
Turbidity	Turbidity meter		APHA 2130
pH	pH meter.	Distilled water	APHA 4500-HB
Colour	DR6000, 100 ml conical flask and pipette		HACH 8025
Ammoniacal-nitrogen	DR6000, pipet, and graduated cylinder	Reagent Nessler, Polyvinyl alcohol, mineral stabilizer, and deionized water	HACH 8038
Dissolved Oxygen (DO)	DO meter and beaker.	Distilled water	APHA 4500-OG
Biochemical Oxygen Demand (BOD)	BOD bottles (300 ml), BOD incubator, pipette	BOD Nutrient buffer pillows (3 ml), distilled water	HACH 8043
Chemical Oxygen Demand (COD)	DRB 200 reactor beaker and pipet.	Sulphuric Acid (H2SO4), Potassium Dichromate (K2Cr2O7), Mercuric Sulphate (HgSO4), and COD digestion vial (high range).	HACH 8000
AEC	Aluminium electrode, Ferum electrode, 1 L beaker, magnetic stirrer, wire, crocodile clip, aeration diffuser, air pump, airflow meter, DC power supply	0.1M (H2SO4),1N sodium hydroxide (NaOH), 40 ml sodium chloride (NaCl)	

2.4 Experimental Work

AEC treatment is a hybrid of physical and chemical methods that integrate aeration into electrochemical coagulation. Before implementing AEC treatment, leachate characterization was performed to analyze its content and concentrations. Then, AEC method was run under two optimizations, pH and electrolysis duration. The volume of leachate sample for each experiment was fixed at 750 ml. A combination of aluminium and ferum electrodes was used in this study with dimensions 200 mm x 70 mm x 1 mm. The electrodes were dipped in the beaker to a depth of 90 mm with the distance between the electrodes fixed at 50 mm. Next, PAC coagulant with a dosage amount of 250 mg/L was added into sample. A digital DC power supply was used to give a regulated electricity current where the current density was fixed at 200 A/mm^2. An air pump with air flow meter was used to provide aeration into the sample, where the aeration rate was constant at 1.0 L/min. The beaker containing the leachate sample was pumped with air from the air pump during the aeration process. The airflow meter was linked to the air pump and will act as a pressure controller. After 30 minutes of settling time, the percentage removal of colour, ammoniacal-nitrogen and suspended solid was measured. The values of each removal parameter's initial and final concentrations were used to calculate the removal percentage using Eq. (1). Fig. 2 illustrates the schematic diagram of AEC experimental setup.

$$\text{Removal} = \frac{(C_0 - C_f)}{C_0} \times 100 \quad (1)$$

where C_0 = Initial concentration of each removal parameter, and C_f = Concentration of each parameter.

3. Results and Discussion

3.1 Leachate Characteristics

The leachate was characterized as old leachate considering pH value greater than 7.5, COD value lower than 4000 mg/L, and BOD5 /COD ratio less than 0.1. From the data, pH 8.18 was obtained, a bit lower than pH 8.6 observed by Anuar et al. [22]. Both values were greater than pH 7.5, therefore categorizing the leachate as old leachate. Meanwhile, the current COD value in this study was 789 mg/L, considered low, thereby characterized as old leachate since the limitation of COD value was below 4000 mg/L for old leachate. According to Zailani et al. [23], the BOD_5/COD ratio was used to determine the biodegradability of leachate, which also indicates the landfill's age. For old leachate, the ratio was less than 0.1. From the ratio, the biodegradability of the leachate was low, at 0.038. Hence, the leachate was characterized as stabilized old leachate. For suspended solid, colour and ammoniacal-nitrogen, it was found that the value was 109 mg/L, 2607 ADMI and 1592 mg/L, respectively. These recorded values were greater than the allowable limit discharge by EQA 1974 [21]. The best treatment method for treating old or stabilized leachate is physical and chemical treatment. Thus, leachate from SRL is suitable to be used for AEC study. Table 2 tabulates the values of leachate characterization parameters from the Simpang Renggam Landfill compared to EQA 2009 [21].

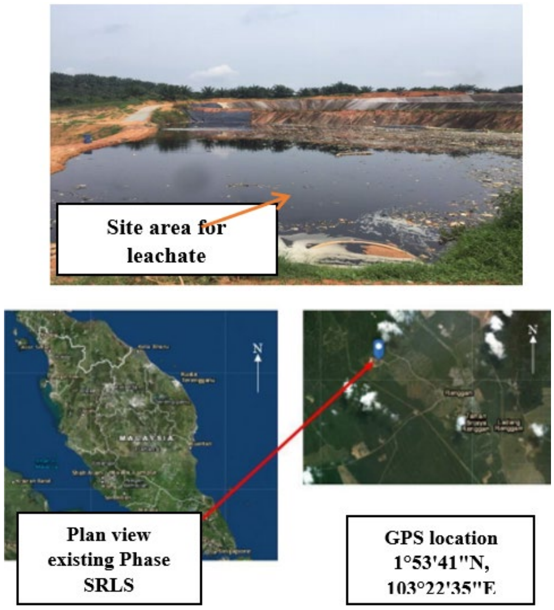

Fig. 1 - Simpang Renggam landfill site

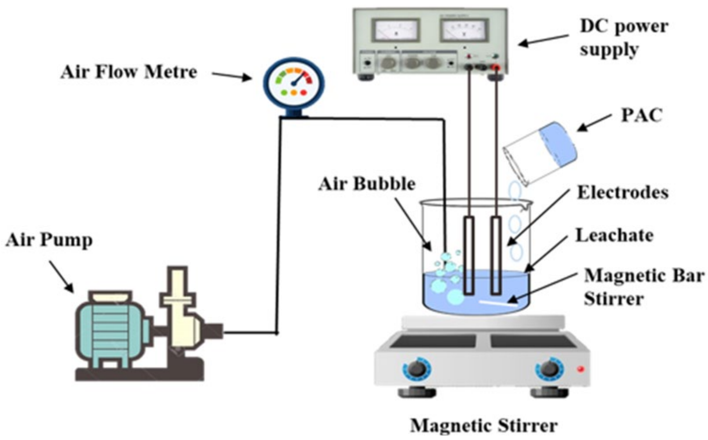

Fig. 2 - Schematic diagram of AEC experimental setup

Table 2 - Characteristic of leachate from Simpang Renggam Landfill compared to EQA 2009

Parameter	Unit	Average	EQA 2009
Turbidity	NTU	35	
Suspended solid	mg/L	1.000	50
pH		8.18	6.0 – 9.0
COD	mg/L	789	400
Colour	ADMI	2607	100
Ammoniacal-nitrogen	mg/L	1592	5
BOD_5	mg/L	30	20

3.2 Optimum pH for AEC

From Fig. 3, suspended solid and colour removal efficiency were seen to be higher in acidic conditions compared to neutral and alkali conditions. The highest removal percentage for suspended solid and colour were recorded at pH 6 and 5. According to Nasrullah et al. [24], the electro-generation of Ferric ions caused high treatment efficiencies were achieved in a relatively acidic medium, 3 to 6 pH value. On the other hand, pH towards acidic value affected ammoniacal-nitrogen removal efficiency poorly, while a better result was achieved when pH was in neutral and alkaline value. The ammoniacal-nitrogen removal percentage peaked at pH 8, with 49%. A previous study by Mahvi et al. [25] stated in strongly alkaline conditions, a strong oxidizing agent, HOC1 changed to chlorate ion, ClO_3^- which has a lower oxidizing potential than HOCl, thereby ammoniacal-nitrogen oxidation rate and removal efficiency is lower in strongly alkaline conditions. Hence, neutral pH is recommended for ammoniacal-nitrogen removal due to higher concentrations of HOCl. Comparing to previous research, it can be summarized that a better result was accomplished when the initial pH was at an acidic value. From analysis, the best optimization for initial pH was at pH 5 because the removal of suspended solid and colour was highly satisfied with 84% and 88%, respectively. However, the removal of ammoniacal-nitrogen was not good since the removal was only 18%.

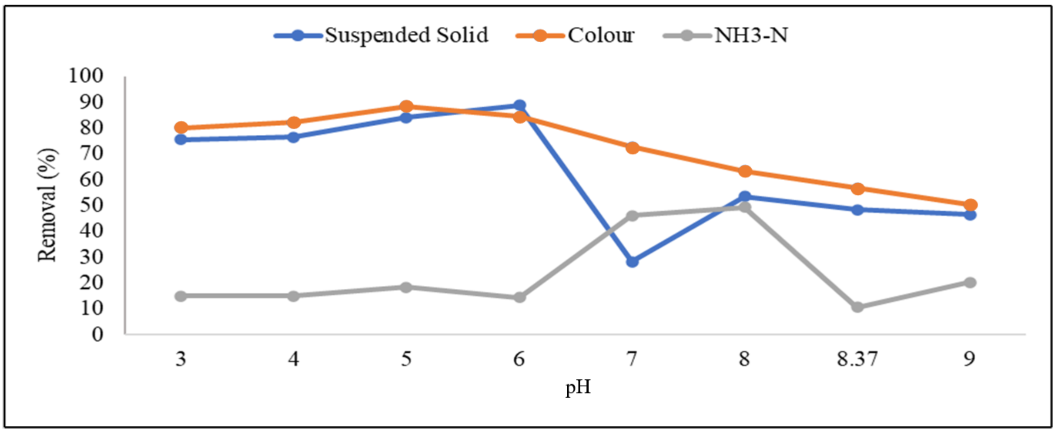

Fig. 3 - Percentage removal of suspended solid, colour and ammoniacal-nitrogen at optimum pH value

3.3 Optimum Electrolysis Duration for AEC

Based on the graph in Fig. 4, the highest removal efficiency of ammoniacal-nitrogen (40%), colour (93%) and suspended solid (93%) was recorded at 10 minutes of electrolysis duration. However, the removal slowly decreased and fluctuated after reaching its optimum electrolysis duration. A similar observation was recorded in the case of suspended solid, colour and ammoniacal-nitrogen removal. In terms of suspended solid, this behavior might be because the EC treatment causes the suspended particles to settle faster, causing more suspended particles to clump together. When wastewater is exposed to EC treatment, the ionic charge increases, causing more particles to collide, eventually aiding particle aggregation and attraction [26]. Moreover, increasing the reaction time may raise the temperature of the wastewater, thereby increasing the gas bubbles' kinetic energy and random motion [27]. The process of pollutant attachment to floc may be hampered by the random motion of gas bubbles and tiny colloidal particles. For ammoniacal-nitrogen, when the electrolysis duration was extended, iron ions created hydroxide flocs, and the bubble production rate increased. As a result of the co-effect of coagulation and flotation, the contaminants in leachate were eliminated [28]. Hence, from the data analysis, it can be determined that the optimum electrolysis duration was at 10 minutes, with the highest removal of ammoniacal-nitrogen, colour and suspended solid. Furthermore, considering economic factors, less energy consumption and shorter duration are able to save the operating cost of treatment.

3.4 Comparison Treated Effluent with Standard Leachate Discharge

The efficiency of AEC in removing suspended solid, colour and ammoniacal-nitrogen from stabilised treated leachate were compared to standard leachate discharge by Environmental Quality (Control of Pollution from Solid Wastewater Transfer Station and Landfill) Regulation 2009 and was presented in Table 3. From Table 3, the standard discharge for the suspended solid was 50 mg/L. The suspended solid in raw leachate was 89 mg/L. After leachate was treated with AEC method, the suspended solid was reduced to 6 mg/L, which complied with the required standard. After the AEC treatment process, the value of colour was reduced from 1971 to 135 ADMI which covered 93% of removal but still did not meet the standard effluent discharge. Similar observation was recorded for ammoniacal-nitrogen, where the treated leachate after AEC treatment only achieved 870 mg/L, whereby the standard of ammoniacal-nitrogen discharge was 5 mg/L. AEC method for treating old leachate was highly effective in removing suspended solid. Overall, only suspended solid complied with the effluent discharge standard. Whereas ammoniacal-nitrogen and colour required further treatment to achieve the permissible limit.

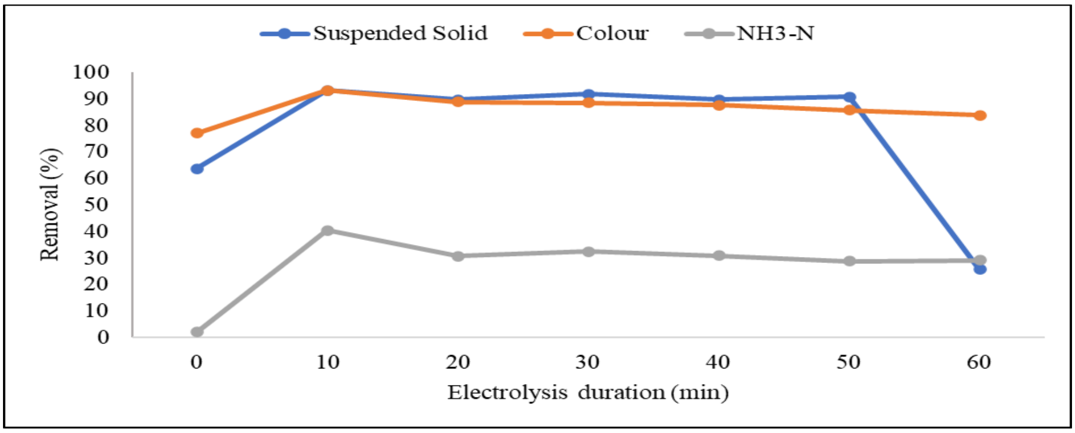

Fig. 4 - Percentage removal of suspended solid, colour and ammoniacal-nitrogen at optimum electrolysis duration, 10 minutes

Table 3 - Characteristic of leachate from Simpang Renggam Landfill compared to EQA 2009

Parameter	Discharge Limit According To Environmental	Untreated leachate	Treated leachate using AEC method
Suspended solid (mg/L)	50	89	6
Colour (ADMI)	100	1971	135
Ammoniacal-nitrogen (mg/L)	5	1460	870

4. Conclusion

The efficiency of leachate treatment using AEC was influenced by electrolysis duration and pH. At 10 minutes of electrolysis duration and pH 5 resulted in the best removal efficiency of suspended solid (93%), colour (93%) and ammoniacal-nitrogen (40%). From observation, the utilization of aeration into EC method in treating leachate has demonstrated a positive effect in removal efficiency. However, colour and ammoniacal-nitrogen still do not meet the acceptable condition of leachate discharge by Environmental Quality (Control of Pollution from Solid Wastewater Transfer Station and Landfill) Regulation 2009. With such preliminary findings, the AEC method can be considered a treatment option to enhance the elimination of suspended solid from leachate.

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