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Treatment of landfill leachate using palm shell-activated carbon column: Axial dispersion modeling and treatment profile

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ABSTRACT

The flow inside an adsorption column for the treatment of landfill leachate is a poorly understood parameter due to lack of studies on its fluid dynamics. In order to address this matter, axial dispersion modelling was conducted to determine the flow pattern of landfill leachate in a column with palm shell-activated carbon (PSAC) as media. In addition, the treatment profile of leachate via adsorption onto PSAC in terms of chemical oxygen demand (COD) and turbidity removal as well as leachate pH was studied. Lithium chloride (LiCl) was used as a non-reactive tracer. The vessel dispersion number (D/uL) was determined to be between 0.01 and 1, implying small and slow dispersion occurring in a plug flow-like dynamics. The model was applicable for modeling of leachate flow inside a pack bed column. Empty bed contact time (EBCT) was utilized to determine the treatment profile with regards to COD and turbidity removal from leachate. The highest 50% breakthrough of COD removal at 1460 mg/g was obtained at EBCT of 14.7 min. The EBCT has no significant effect on turbidity removal and effluent pH.

1. Introduction

It is a well-known fact that leachate originating from a municipal wastes landfill is a threat to surface and groundwater. Landfill leachate contains high amounts of organics, inorganics and metals and has high concentration of chemical oxygen demand (COD) which is detrimental to the survival of aquatic lifeform. A wide spectrum of treatment technologies has been utilized for treating landfill leachate. Examples include precipitation, coagulation, membrane separation, ion exchange, bioadsorption, adsorption and ozonation [1]. Adsorption using activated carbon is also a well-recognized means of leachate treatment. Activated carbon is considered as one of the most effective adsorbents, especially for substances containing refractory organic compounds that resist biodegradation and persist in the environment [2]. It can be used as a standalone treatment method [3] as well as in tandem with biological [4–6], ozonation [7] and limestone-based [8] methods. In general, the application of activated carbon adsorption is effective for the removal of non-biodegradable compounds from landfill leachate [9].

While the adsorptive mechanisms of solutes onto surface of activated carbon are fairly established, nonetheless, the fluid dynamics and mechanisms inside such activated carbon columns for treatment of landfill leachate are poorly understood. In reality, the flow inside such columns is non-ideal in which a portion of the flow that enters the reactor during a given time period arrives at the outlet before the bulk of the flow that entered the reactor during the same period arrives [1]. The first step in addressing this problem is to model the flow inside the column in order to provide an understanding of its type of flow. This is necessary for column modification to improve its performance. An example of such model is the axial dispersion model. It is derived from modification of ideal plug flow of a fluid, which is taking consideration into some degree of backmixing where the magnitude is independent of position within the reactor [10]. The main objectives of this study were (a) to conduct an axial dispersion modeling study via a non-reactive tracer to determine the flow pattern of column bed, hydraulic retention time (HRT) and effective volume of the column bed as well as; (b) to analyze the leachate treatment profile by adsorption using palm shell-activated carbon (PSAC) column.

2. Experimental

2.1. Collection and characterization of landfill leachate

Landfill leachate samples were collected from the inlet feed to a leachate treatment facility located in a landfill at Puchong, Selangor, Malaysia. The samples were collected via grab sampling in

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Table 1
Chemical characteristics of landfill leachate

Parameter	
pH	7.7
Temperature (°C)	26.3
Chemical oxygen demand (COD) (mg/L)	10800.0
Biochemical oxygen demand (BOD) (mg/L)	5400.0
Total suspended solids (mg/L)	900.0
Turbidity (NTU)	690.0
Color (hazen)	400.0
Dissolved oxygen (mg/L)	0.03
Total Kjeldahl nitrogen (mg/L)	2000.0
Ammonical nitrogen (mg/L)	2900.0
Phosphate (mg/L)	190.0
Iron (mg/L)	31.7
Manganese (mg/L)	0.2
Nickel (mg/L)	1.3
Zinc (mg/L)	1.4
Cadmium (mg/L)	0.3

clean polyethylene bottles. Characterization of the leachate was conducted five times using standard methods stipulated in APHA [11] and the average result is shown in Table 1. COD concentrations were determined using COD reactor from Cole-Palmer. pH measurements were conducted using Metrohm 654 pH meter from Schmidt Scientific. Turbidity was analyzed using Photometer SQ118 from Merck. Color was determined using Lovibond 2000 comparator. Prior to determination of phosphorus concentrations, digestion of the leachate was conducted using APHA Method 4500 [11]. Phosphorus concentrations were measured via 150-20 spectrophotometer from Hitachi. Heavy metals concentrations were analyzed via PerkinElmer Optima 3000 inductively coupled plasma-optical emission spectrometer (ICP-OES).

2.2. Palm shell-activated carbon

PSAC derived from palm shell was obtained from Kekwa Indah Sdn. Bhd., a local producer located in Negeri Sembilan, Malaysia. It was ground and sieved to produce particles of sizes ranging from 0.6 to 1.18 mm. It was then washed thoroughly with distilled water to remove impurities and unwanted moisture. Subsequently, the PSAC was dried in an oven at 103 °C overnight, cooled at room temperature and finally stored in a desiccator. The surface area and pore volume of the PSAC were determined to be 595 m^2/g and 0.36 cm^3/g, respectively in a study conducted by Leong [12] using Micrometrics ASAP 2010 surface area analyzer. Prior to analysis, the samples were degassed at 120 °C for at least 24 h.

2.3. Dispersion modeling study

The modeling study was conducted using a downflow column with an activated carbon as media (height = 40 cm; i.d. = 5 cm; fixed bed height = 30 cm). Lithium chloride (LiCl) was used as a non-reactive tracer for this study [10]. The tracer method used in this study was similar to the method specified by Levenspiel [10] and Levenspiel and Smith [13]. An injector with 10 mL syringe was used to inject LiCl solution into the influent line of column bed with constant velocity of 80 mL/min. Samples were collected at the column effluent at 1-min interval and analyzed for Li^+ concentration using ICP-OES. All chemicals used in this study were reagent grade and obtained from Merck (Malaysia).

2.4. Fixed bed adsorption study

Three activated carbon columns designed in series with column and packing dimensions similar to the modeling study were used for the treatment of landfill leachate. Leachate was pumped to the top of the first column using a Masterflex peristaltic pump at flow rate of 40 mL/min and subsequently flowed to the second and third columns. The retention time in each column is 10 min. The approximate bed volume of each column was 590 L. The pH leachate was adjusted to 7 using sulfuric acid before inlet flow. The study was conducted at temperature 25 °C. Samples were taken from the column effluents at different time intervals and analyzed for pH, COD and turbidity.

3. Results and discussion

3.1. Dispersion modeling study

Fig. 1 shows the concentration profile of effluent Li^+ throughout the modeling period. It is obvious from the curve that it is not symmetrical. This indicates large deviation from plug flow which provides an initial indication that D/uL value for this fixed bed column is more than 0.01.

For the axial dispersion model, vessel dispersion number (D/uL), a dimensionless group, is used to measure the extent of axial dispersion where D is the axial dispersion coefficient (m^2/s), u the velocity of fluid (m/s) and L the height of the vessel. If D/uL is close to zero, then dispersion in the vessel is negligible and plug flow is deemed to occur throughout the vessel whereas if D/uL is close to infinity, then there is large dispersion/mixed flow in the vessel. For closed vessels and non-symmetrical tracer curve, D/uL can be determined from the following equation:

$$\sigma_\theta^2 = \frac{\sigma^2}{\bar{t}^2} = 2\left(\frac{D}{uL}\right) - 2\left(\frac{D}{uL}\right)^2 [1 - e^{-uL/D}] \tag{1}$$

where $\bar{t}$ is the mean time of passage/measured HRT (min) and σ^2 is the variance. The $\bar{t}$ and σ^2 parameters can be calculated via the following equations [10]:

$$\bar{t} = \frac{\Sigma t_i C_i}{\Sigma C_i} \tag{2}$$

$$\sigma^2 = \frac{\Sigma t_i^2 C_i}{\Sigma C_i} - \bar{t}^2 \tag{3}$$

where C is concentration of Li^+ (mg/L) at time t (min).

Table 2 lists the parameters obtained from the axial dispersion model. The percentage loss of tracer was calculated based on simple mass balance calculation. The percentage loss of tracer is marginal in this study (<1.5%), indicating steady-state condition in the study in which accumulation of Li^+ in the column is negligible. The obtained D/uL value is higher than 0.01, thus confirming the validity of the tracer study. Levenspiel [10] suggests that D/uL should not be higher than 1 in order for the dispersion model to be

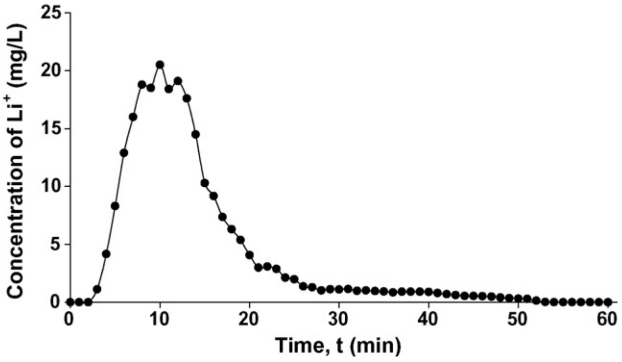

Fig. 1. Concentration profile of effluent Li^+ throughout the modeling period.

Table 2
Parameters obtained from axial dispersion model

$\bar{t}$ (min)	13.85
$t_{\mathrm{theoretical}}$ (min)	10
σ_θ^2 (min^2)	0.3638
D/uL	0.2375
Volume, V (L)	0.8
Effective volume, V_e (L)	1.11
Loss of tracer (%)	1.35

Table 3
Effect of EBCT on column performance at 50% breakthrough

EBCT (min)	Bed depth (cm)	Volume treated (L)	Service time (min)	$Q_{0.5}$ (mg/g)
14.7	30	72	1800	1460
29.4	60	85	2125	638
44.1	90	162	4050	847

valid. Therefore, since D/uL for our study is lower than 1, the model is applicable. The measured HRT of the tracer for the column bed (13.85 min) is observed to be higher than the theoretical HRT of 10 min. This suggests non-existence or very marginal occurrence of dead space or stagnant pockets in the column. This notion is further reinforced by higher effective volume as compared to theoretical volume. The overall modeling result indicates small and slow dispersion occurring in a plug flow-like dynamics (i.e. marginal deviation from plug flow).

3.2. Fixed bed adsorption study

Empty bed contact time (EBCT) is defined as the time a fluid spends in a column, on the basis that the column is empty [14]. EBCT can be expressed as follows:

$$\mathrm{EBCT\ (min)} = \frac{\mathrm{bed\ \ volume\,(L)}}{\mathrm{volumetric\ \ flow\ \ rate\,(L/min)}} \tag{4}$$

Each column had EBCT of approximately 14.7 min and a total of 44.1 min of contact time in the three columns. Fig. 2 shows the breakthrough curves of COD removal using the activated carbon columns. Even though the curves exhibit an S-shape, they are not really well-defined as fluctuations occur in the COD removal process. This is most probably attributed to an amalgamation of solutes (inorganics and organics) which render the activated carbon adsorption more complex. This phenomenon was also observed in a study conducted by Morawe et al. [3]. For the curves, the effluent concentrations never reach the influent COD concentration. This implies a possibility of biological activity in the columns in which the activated carbon may act as support for degrading bacteria already present in the leachate. This observation was also reported by Morawe et al. [3].

In order to investigate the impact of EBCT on COD removal, 50% breakthrough was taken as a criterion [15]. Table 3 shows the effect of EBCT on column performance at 50% breakthrough. The increase in EBCT from 14.7 to 44.1 min results in an increase of 125% in volume treated at 50% breakthrough. It is also observed that increase in bed depth results in increase of service time in an exponential manner. The 50% breakthrough capacity, $Q_{0.5}$ (mg/g) can be calculated using the following equation [16]:

$$Q_{0.5} = \frac{C_e V_{\mathrm{mb}}}{w} \tag{5}$$

where V_{mb} is the volume at 50% breakthrough (L), w the weight of the activated carbon (g) and C_e the concentration of the effluent at 50% breakthrough (mg/L). It was determined that the highest $Q_{0.5}$ of 1460 mg/g was obtained at EBCT of 14.7 min. This implies that 14.7 min of contact time is adequate for COD removal during the continuous column operation.

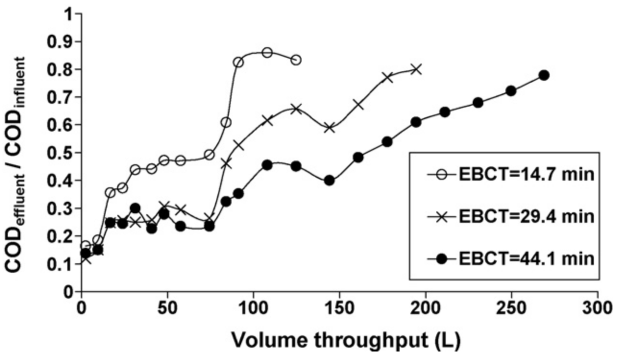

Fig. 2. Breakthrough curves of COD removal using the activated carbon columns.

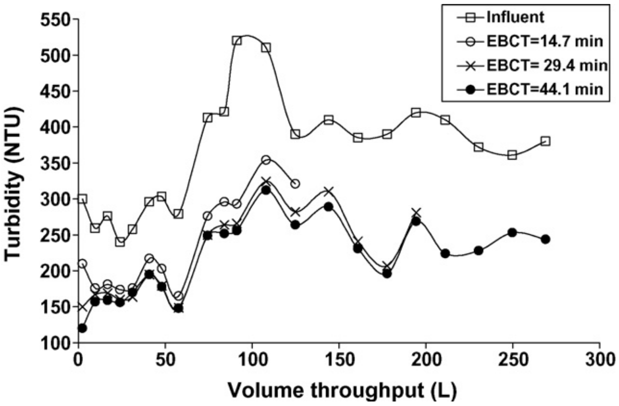

Fig. 3. Turbidity profile of leachate.

Fig. 3 shows the turbidity profile of leachate at corresponding volume throughput. The turbidity removal percentages at EBCT of 14.9, 29.4 and 44.1 min range from 17.7 to 40.9%, 24.4 to 50.0% and 30.0 to 60.0%, respectively. It is evident that the EBCT has no significant effect on turbidity removal since only marginal difference on turbidity values was observed for all three EBCTs. This indicates that at EBCT of 14.7 min, turbidity removal is almost at the maximum and that increases in EBCT only marginally increase turbidity removal.

Fig. 4 shows the profile of leachate pH at corresponding volume throughput. Marginal fluctuations in the profile (<1) are observed. Effluent pH values are approximately 0.12–0.67 units higher than

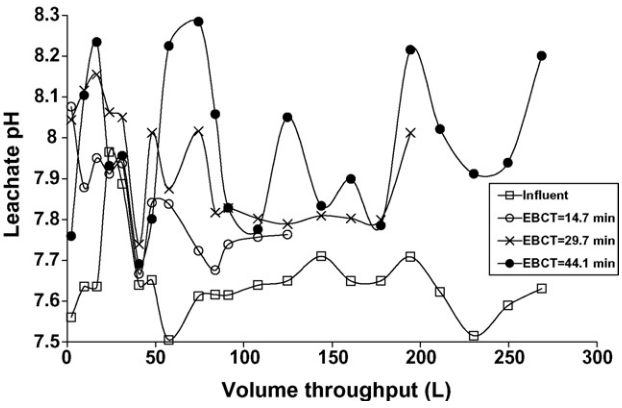

Fig. 4. Profile of leachate pH.

influent pH. The increase in pH of leachate after contact with activated carbon is most probably due to adsorption of H^+ ions on surface of activated carbon as palm shell-activated carbon is known to contain a sizeable amount of acidic functional groups such as lactonic and carboxylic acids [17]. This results in relative reduction of H^+ ions increase of OH^- ions in the bulk solution. The EBCT has no significant effect on effluent pH since only marginal difference on pH values was observed for all three EBCTs.

4. Conclusions

The vessel dispersion number (D/uL) was determined to be in between 0.01 and 1, suggesting small and slow dispersion occurring in a plug flow-like dynamics (i.e. marginal deviation from plug flow. The axial dispersion model was applicable for modeling of leachate flow inside a pack bed column. The highest 50% breakthrough of COD removal at 1460 mg/g was obtained at EBCT of 14.7 min. The EBCT has no significant effect on turbidity removal and effluent pH.

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