Removal of High-Strength Colour from Semi-Aerobic Stabilized Landfill Leachate via Adsorption on Limestone and Activated Carbon Mixture

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Abstract

This study was undertaken to investigate the suitability of using a mixture of activated carbon (AC) and limestone (LS) as an adsorbate medium for removing high-strength colour from a semi-aerobic stabilized landfill leachate. Colour adsorption was explained by the Langmuir and Freundlich isotherms. Moreover, the adsorption kinetic mechanism was investigated using pseudo-first-order and second-order kinetics models. Approximately 88% of colour removal was achieved using a mixture of LS ($35cm^3$) and AC ($5cm^3$). In the current study, the Freundlich and Langmuir isotherms offered very good fits for colour adsorption with R^2 values of more than 0.96. The empirical constant (n) value of the Freundlich isotherm was greater than 1, which implies a beneficial adsorption for colour. Results obtained from the kinetic models showed good compliance with the pseudo-second-order equation ($R^2 = 0.997$). This indicates that LS can be used as a cost-effective medium to replace AC for colour removal at a considerably lower cost.

Keywords: Semi-aerobic landfill, stabilized leachate, adsorption, activated carbon, limestone, kinetics.

Introduction

Landfills have served as the ultimate solid waste disposal mechanism in most of the countries[1,2,3]. The generation of leachate is one of the main concerns associated with landfills and it is required to be properly collected and treated to reduce negative environmental impact on nearby water resources through its percolation[4]. Landfill leachate is a complex fluid with considerable variations in both quantity and composition[5]. Leachate is produced when moisture combines with refuse in a landfill, extracts contaminants into the liquid phase and produces moisture content sufficiently high to initiate liquid flow. It has been well established that landfill leachate composition depends on many factors, including waste composition, landfill age, landfill design and operation, availability of moisture and oxygen and site hydrology[6,7].

Colour is a common pollutant in landfill leachate[8]. In general, leachate produced by an old landfill with low biodegradability ($BOD_5/COD < 0.1$) is classified as a stabilized leachate. Stabilized leachate classically contains high levels of organic substances such as humic and fluvic compounds, which can be indicated by leachate colour[9]. Humic substances are natural organic matter made up of complex structures of polymerized organic acids, carboxylic acids and carbohydrates[10]. Colour formation in leachate also depends on the types of waste deposited on site. Paints, pigments, colour compounds and iron-based materials, such as those used in construction, commonly constitute colour.

Biological processes are generally effective for new leachates containing mainly volatile fatty acids, but less so for stabilized ones[10]. However, due to its complicated characteristics, stabilized leachate is difficult to treat biologically. Nevertheless, various techniques have been applied to remove colour from stabilized leachate with varying degrees of success. These techniques include oxidation[11,12], flocculation coagulation[8,13], ion exchange[8,14], dissolved air flotation[15] adsorption[16,17].

There has been a tendency to use low-cost available materials for the adsorption of colour from different kinds of wastewater and aqueous solutions in the last two decades. A wide variety of materials have been used as low-cost alternatives to activated carbon (AC) [18,19,20,21]. Most applications for these materials are focused toward industrial wastewater.

Aziz et al.[22,23] have demonstrated the use of limestone (LS) as a low-cost adsorbent. Foul et al.[17] also examined the effectiveness of LS in removing colour from anaerobic leachate. The present study focuses on the removal of high strength colour from semi-aerobic landfill stabilized leachate generated from Pulau Burung Landfill Site (PBLS) by using a mixture of AC and LS.

The landfill site is situated within Byram Forest Reserve at 5º 24' N, 100º 24'E in Penang, Malaysia. This site has a natural marine clay liner. The site receives approximately 2,200 tons of solid wastes daily. PBLS has a semi-aerobic system and is one of only three sites of its kind in Malaysia[23]. A convection process takes place in a semi-aerobic system whereby the decomposition of organic matter inside the landfill causes a rise in temperature. The difference in temperature between the outside and inside of the landfill generates a convection current that goes into the landfill through the leachate pipe[23].

This site experiences highly coloured and turbid leachate due to the presence of a combination of organic matters, suspended solids turbidity[8]. Therefore, the main objective of the present study is to investigate the suitability of using a mixture of AC and LS as a low-cost adsorbent in removing colour from semi-aerobic stabilized landfill leachate. The results will be used as a basis for the search of a cheaper alternative adsorbent for this type of landfill sites. Equilibrium isotherms in this study are analyzed and described using the Langmuir and Freundlich isotherms. Experimental data using the pseudo-first-order and second-order adsorption kinetic models are also determined.

Material and Methods

Media properties: Prior to the experiments, a known volume of LS and AC was air-dried for 24 h. The chemical composition of LS (table 1) was determined via digestion method with concentrated hydrochloric acid (HCl), followed by metal determination with an atomic spectrophotometer (Model Shimadzu AA 660), as detailed in Standard Method for the Examination of Water and Wastewater[24]. The LS used in the present study contained 96.5% $CaCO_3$ and 2.5% $MgCO_3$ (table 1). AC used in the study was commercially available. The density for each media was determined according to the American Society for Testing and Materials standards (ASTM) [25]. The Brunauer-Emmett-Teller-(BET) method, which uses Micromeritics Accelerated Surface Area and Porosimetry Analysers (ASAP) 2000, was employed to estimate the specific surface area. LS was sieved to obtain a size range between 2.36 to 4.75 mm (i.e. the size suggested elsewhere)[22]. The physical properties of the AC and LC are given in table 2.

Table-1
Chemical compositions of limestone

Compositions	Percentage (%)
$CaCO_3$	96.5
$MgCO_3$	2.5
Others	1.0
Total	100

Table-2
Physical properties of activated carbon and limestone

Properties	Activated carbon (AC)	Limestone (LS)
Grade	KI5060	--------
Particle Size (mm)	2.36 - 4.75	2.36- 4.75
Iodine number (mg/g. min)	590 .22	241.17
pH	9-10	-----
Ash Content (%)	5	-----
Surface area (m^2/g)	1786.2	4.386
Particle density (kg/m^3)	1116	2491.16
Bulk density (kg/m^3)	610	1410
Void ratio (%)	45.3	43.4

Leachate sampling characteristics: The sampling method for leachate from PBLS was in accordance with procedures described in Standard Methods for the Examination of Water and Wastewater[24]. Leachate was obtained manually from a pond at an approximate depth of 0.3 m. The samples were immediately transported to the laboratory and stored in a cold room at 4°C prior to experimental use to minimize biological and chemical reactions. The characteristics of the semi-aerobic leachate are illustrated in table 3.

Table-3
The characteristics of raw leachate from an old detention pond at PBLS

Parameter	Values*
BOD_5 (mg/L)	50-150
COD (mg/L)	1533-3500
Suspended Solid (mg/L)	159-233
Turbidity (NTU)	61-198
pH	7.5-9.4
Zinc (mg/L)	0.1-0.6
Iron (mg/L)	1.5-5.2
Copper (mg/L)	4.6-5.3
Chromium (VI) (mg/L)	0.3-0.6
Colour (Platinum unit, PtCo)	3,113-6,180

*Base on 16 month reading

Batch study: A batch study was conducted to establish the removal pattern of colour using the LS and AC mixture (LS:AC). LS were used as a replacement material for AC because of its lower cost. The batch equilibrium experiments were conducted by mixing a fixed amount of media (measured in terms of volume, 40 cm^3) with 120 mL of raw leachate sample in a 250 mL conical flask. The influence of shaking speed, shaking time settling time was pre-examined prior to the main experiments. The former was conducted by varying the shaking speed of the orbital shaker between 50 and 350 rpm (model Bioblock Scientific Agitator 74578). The mixture was kept at a mixing ratio of 35:5 (LS:AC) by volume. Shaking time was 60 min and the samples were allowed to settle for 90 min before decanting for colour determination[23]. The effect of shaking time was determined by repeating the experiment using a determined shaking speed and a settling time of 90 min. The experiment was further repeated to determine the settling time, a factor which may influence the colour removal. This was performed by allowing samples to settle at different times (between 10 and 120 min) using results of both shaking speed and shaking time. Later, pre-determined results were used in the main experiments, in which different mixture ratios of LS to AC were used for specific volumes of leachate. The effect of initial colour concentration was also examined at different mixture ratios.

Equilibrium and isotherm models: The amount of adsorption at an equilibrium state, q_e (PtCo/g), was calculated using the following equation:

$$q_e = \frac{(C_o - C_e).V}{W} \qquad ....(1)$$

where C_o and C_e (PtCo/L) are the liquid-phase concentrations of the sample at initial stage and equilibrium, respectively; W(g) is the mass of composite media used; and V(L) is the volume of the solution. Removal efficiency of the parameters studied was calculated as follows:

$$\text{Removal (\%)} = \frac{C_o - C_e}{C_o} x100 \qquad ....(2)$$

Adsorption isotherm is fundamentally necessary to explain how solutes interact with adsorbents is therefore critical to the optimal use of adsorbents[26]. The Langmuir and Freundlich isotherm models are employed in this study. The linearized forms of the two isotherms are as follows:

$$\frac{1}{q_e} = \frac{1}{K_a q_m C_e} + \frac{1}{q_m} \qquad ....(3)$$

$$\log \ q_e = \log \ K_F + \frac{1}{n} \log \ C_e \qquad ....(4)$$

The Langmuir constants, q_m (PtCo/g) and K_a (L/ PtCo), are related to adsorption capacity and energy of adsorption, respectively, while K_F and n are Freundlich constants.

Continuous Flow Experiment: Continuous flow experiments were carried out using only LS:AC (35:5). Perspex filter columns with 8 cm diameter and 30 cm length were used in the current study. The column set-up was cleaned with diluted nitric acid and rinsed with distilled water. Before the start of the experiment, the media was immersed in distilled water for 24 h, followed by the pumping of distilled water in an up-flow direction for 60 min to remove any trapped air from the beds. The leachate was pumped in an upward direction to avoid channeling, at a constant flow rate of 20 mL/min. Colour concentration in the effluent was measured by collecting 5 mL of the effluent at various times.

Analytical Methods: Colour concentration was measured by a DR 2000 HACH spectrophotometer adapted from method No. 2120C in standard method for the examination of water and wastewater[24]. Colour measurements were reported as true colour values using the platinum-cobalt (Pt-Co) method. The sample was filtered using 0.45 µm filter paper before each measurement.

Results and Discussion

Figure 1 shows the results of the preliminary experiment for determining optimum shaking speed, shaking time settling time. Over 95% of the colour removal was achieved at a shaking speed of 350 rpm (figure 1a). At a high agitation speed of 350 rpm, high colour removal was obtained at 60 min of shaking time (figure 1b). According to the results, colour removal rate increased with increasing shaking time, achieving equilibrium at 60 min and remaining constant thereafter. Kadirvelu et al. [27] reported similar results whereby colour removal rate increased with shaking time. Optimum settling time was 90 min (figure 1c). Rigorous mixing and longer solid-liquid contact, which causes better interaction between media and leachate, leads to better removal efficiency at higher agitation speed and longer shaking time. A larger active surface area of media is exposed as shaking continues. The settling time of 90 min obtained in this study matches the value reported by Aziz et al. [22].
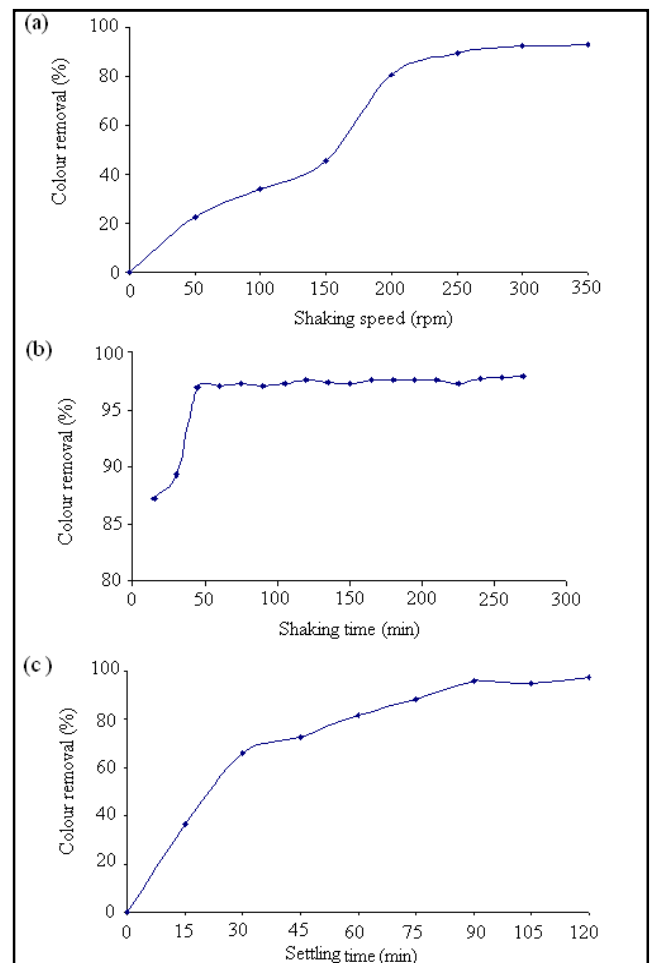
Figure-1
Influence of (a) shaking speed (AC: LS =35:5; shaking time = 1hr; settling time = 1.5hr), (b) shaking time (AC: LS =35:5; shaking speed= 350 rpm; settling time= 1.5hr) (c) settling time on colour removal efficiency (AC: LS =35:5; shaking speed= 350 rpm; shaking time = 1.5hr)

The influence of different LS:AC mixture ratio: Figure 2 shows colour removal efficiency versus LS:AC ratios. More than 80% of colour with an intensity of up to 6,180 Pt Co was removed from the stabilized landfill leachate with AC as the sole media. For a 40 cm^3 quantity of AC or a mixture ratio of 35:5 (LS:AC), the removal rate was about the same. Thus, this mixture rate provides a cheaper

solution with about the same results. In other words, the removal rate of colour was about the same (88%), when AC and its mixture with LS at a ratio of LS:AC = 5:35 was used,. However, the use of LS alone (mixture ratio 40:0) did not produce satisfactory results with less than 40% colour removal.
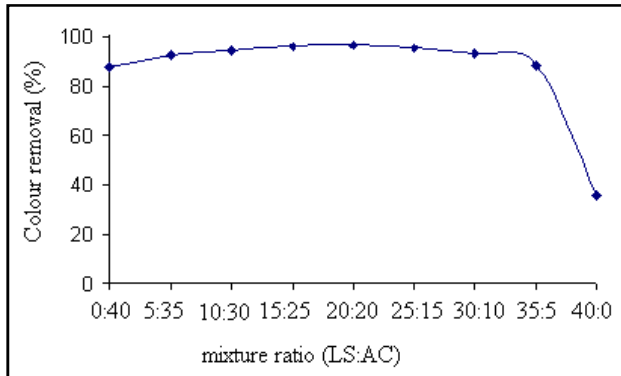

Figure-2
Effect of mixture ratio (LS: AC) on colour removal efficiency (shaking speed = 350 rpm; shaking time = 1hr; settling time = 1.5 hr)

The influence of initial concentration: The effect of colour concentration in the influent on its overall removal efficiency is shown in figure 3. The result of colour removal at a lower initial concentration of colour intensity (50% dilution with initial colour density 6180) gave a slightly better result compared with the results obtained at higher initial concentrations of colour (figure 3). For example, at a mixing ratio of 0:40 (LS:AC), the colour removal was 5% higher (93.4%) at lower initial colour concentration compared to raw leachate without dilution (88%).
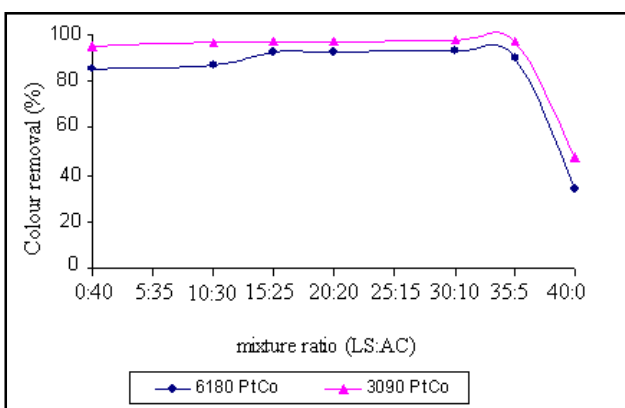

Figure-3
Influence of initial colour concentration on its removal efficiency at different mixture ratio of LS: AC (shaking speed = 350 rpm; shaking time = 1hr; settling time = 1.5 hr)

Adsorption Isotherms: In this study, the Freundlich and Langmuir adsorption models, which have been successfully applied to many adsorption processes[18], were used to study the media adsorption behavior for colour removvla from stabilized landfill leachate. Figure 4 demonstrates the linear plot of the Langmuir and

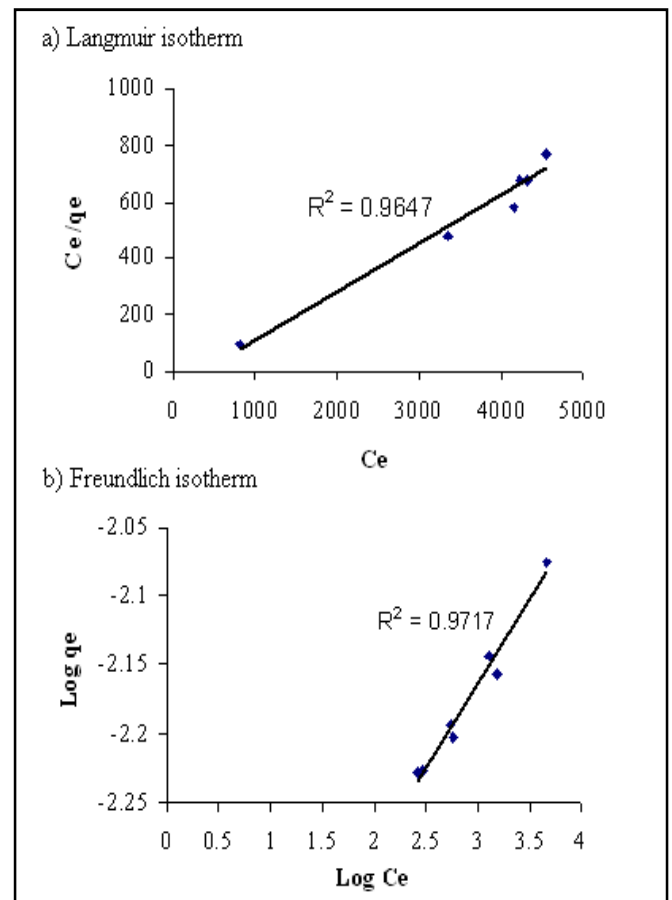Freundlich isotherms for colour adsorption via the LS:AC (35:5) mixture.

Figure-4
The Langmuir and Freundlich isotherms for colour adsorption onto mixture media (LS: AC = 35:5)

The empirical constant values of the Langmuir and Freundlich isotherms for colour were calculated from the linear plot (table 4). The data indicates that both the Langmuir and Freundlich isotherms presented good fits for all studied parameters (R^2>0.95). According to the results, the R^2 value for colour adsorption did not demonstrate any considerable difference between the Langmuir and Freundlich isotherms. Both isotherms could thus be used for colour adsorption using composite media. The empirical constant (n) values of the Freundlich isotherm were greater than 1, implying valuable adsorption for colour from stabilized landfill leachate. Apart from that, the adsorption by mixture media was also favorable for colour, whereby new adsorption sites became available and the adsorption capacity increased with $1/n < 1$.

Adsorption kinetics: Experimental efficiency was determined by the kinetics of adsorption. Several kinetic models are available for predicting the mechanisms involved in the adsorption process[15,23,26]. In the present study, the adsorption kinetics of colour into the LS:AC mixture was investigated using two common models: the pseudo-first-order and pseudo-second-order models.

Table-4
Isotherms equation parameters for colour adsorption onto mixture media (LS:AC = 5:35)

Langmuir isotherm model			Freundlich isotherm model		
q (PtCo/g)	b	R^2	K PtCo/g (L/mg)1/n	1/n	R^2
5.8	0.00275	0.9647	0.0032	0.122	0.9717

Table-5
Kinetic model parameters for colour adsorption onto mixture media (LS:AC = 5:35)

Exp	Pseudo-First –Order Model			Pseudo-Second Order Model		
$q_{e, exp}$ (g/ PtCo)	K_1 (min^{-1})	$q_{e, cal}$ (g/ PtCo)	R^2	K_2 (g/ PtCo .min)	$q_{e, cal}$ (g/PtCo)	R^2
381.5	0.0115	36.48	0.9861	0.00076	384.6	0.9997

The pseudo-first-order model is described as below:

$$\log(q_e - q_t) = \log(q_e) - \frac{k_1 t}{2.303} \qquad \text{..............(6)}$$

The pseudo-second-order model is described as below:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \qquad \text{..............(7)}$$

where q_e and q_t are the amounts of pollutants adsorbed at equilibrium (PtCo/g for colour) and at time t, respectively; and k_1 (min^{-1}) and k_2 (g/PtCo min) are the equilibrium rate constants of the pseudo-first-order and pseudo-second-order models, respectively.
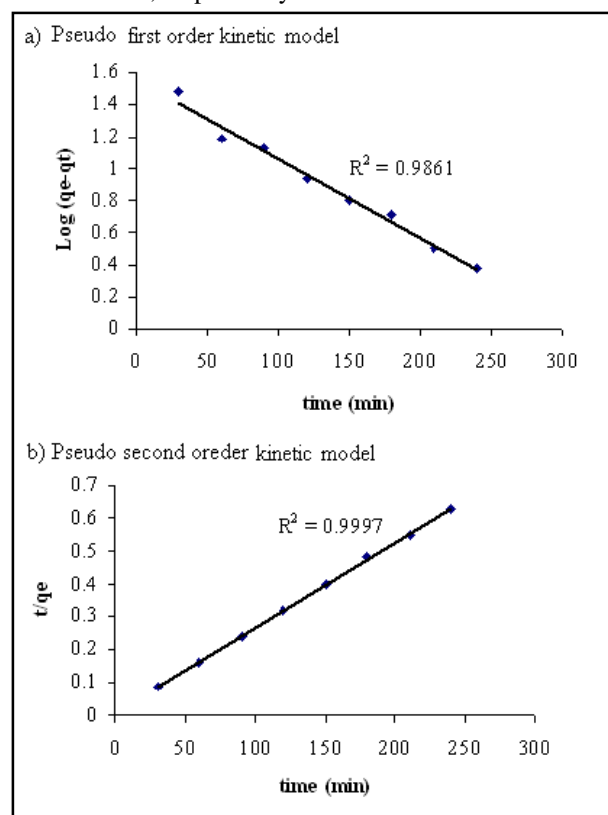

Figure- 5
Pseudo-first-order kinetic model and Pseudo-second-order kinetic model for colour adsorption onto mixture media (LS: AC = 35:5)

Figure 5 demonstrates the linear plots of the pseudo-first-order and pseudo-second-order models. Table 5 shows the values of k_1, k_2, q_e R^2 for the models. In table 5, the correlation coefficient (R^2) for the pseudo-second-order kinetic model is extremely high and close to unity, which is contradictory to both first-order-kinetic models.

The high correlation coefficient (R^2) values indicate that the model successfully describes the adsorption kinetics of the parameters. Furthermore, equilibrium sorption capacities (q_e) calculated from pseudo-second-order models were in good agreement with the experimental values. Consequently, sorption reaction by the pseudo-second-order kinetic model was more favorable, indicating that the adsorption process for colour and COD was controlled by chemisorption[18].

Continuous Flow Studies: An LS:AC mixture in the ratio of 35:5 (V/V) was used for the column study. Figure 6 demonstrates the plot of the percent of colour removal versus time and shows that the column was exhausted within the first 60 min of the experiment for colour removal. The removal rate can reach up to 60%. Limestone can thus be used as a cost-effective medium to replace activated carbon for colour removal at a considerably low cost.
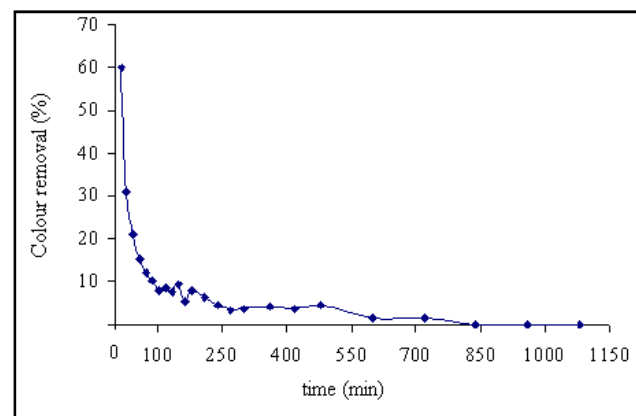

Figure-6
Continuous flow experiment using an LS: AC mixture of 35:5(Q= 20mL/min)

Conclusions

In the present study, about 88% of colour with an initial concentration of 6,180 PtCo was successfully removed from semi-aerobic stabilized landfill leachate using a mixture of LS:AC (35:5), where 35 cm^3 of LS and 5 cm^3 of AC were used. Results also indicated that the removal rate of colour when pure AC (LS:AC = 0:40) was used was almost similar to the removal rate when LS:AC ratio of 35:5 was used. However, the use of LS alone (LS:AC = 40:0) did not yield satisfactory results, showing only about 40% removal. The equilibrium data were well-fitted to the Langmuir and Freundlich isotherm models. Results indicated that both isotherm models were adequate for describing colour removal using composite media because there was no considerable difference between them and the LS+AC mixture. This was confirmed by the R^2 value of each model (which were both >0.96). Results from each kinetic model showed good conformity with the pseudo-second-order equation, demonstrating that the rate of sorption reaction was controlled by the second-order mechanism (chemical sorption). The study demonstrated that the composite media (LS and AC) could be effectively used as a valuable and effective adsorbate for colour reduction from semi-aerobic stabilized landfill leachate.

Acknowledgements

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