Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2019.24247

160 (2019) 185–192
August

Equilibrium isotherm and kinetic study of the adsorption of organic pollutants of leachate by using micro peat-activated carbon composite media

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Received 28 November 2018; Accepted 8 April 2019

ABSTRACT

The suitability of micro peat (MP) and activated carbon (AC) mixed media as a composite adsorbent for the landfill leachate treatment in terms of COD reduction and color removal was investigated in this study. All the adsorption media was pulverized and sieved to a particle size of 150 µm. The MP-AC composite media efficiently removed 87% COD at optimal ratio 1.5:2.5 and 74% color at optimal ratio 2.0:2.0, within 120 min of contact time, 200 rpm of shaking speed at pH 7. The findings revealed that Langmuir isotherm best described the uptake of COD and color which implies that the adsorption of leachate in this study onto MP-AC composite media was homogeneous on a mono layer surface. The kinetic data and the factor controlling adsorption process fitted better to the pseudo-second-order model. Energy of adsorption (E) were observed as 267.26 kJ mol^{-1} and 79.06 kJ mol^{-1} for COD and color respectively indicating chemisorption as the limiting controlling step for the reduction of COD and color onto MP-AC composite media. The overall rate of the COD and color adsorption processes onto MP-AC composite media appears to be dominated by chemisorption. The pattern of adsorption showed that micro peat offers a cost effective alternative and can be utilized as partial replacement of conventional media (AC) for the removal of organic matters from landfill leachate.

Keywords: Activated carbon (AC); Composite; COD; Color; Leachate; Micro peat (MP)

1. Introduction

Landfill leachate is a complex wastewater produced as a result of the interaction of waste deposit with water percolating through the body of a landfill. The wastewater contains high load of organic matter, ammoniacal nitrogen, inorganic salts, chlorinated organic, and heavy metals content [1]. Additionally, the composition of leachate is highly site specific and varies widely depending on many factors such as the waste composition, site hydrology, the availability of moisture and oxygen, design and operation of the landfill, and its age [2]. Furthermore, it is also reported that the organic matter present in the leachate or other wastewater act as major contaminants as it has tendency to pollute water bodies and surrounding land [3,4]. In addition, wastewater with organic contaminants contain large quantities of suspended solid that may generate hydrogen which contributes majorly to greenhouse emission of gasses [5,6]. Hence, it becomes necessary to treat landfill leachate or wastewater before it is discharged into water bodies and land to ensure environmental sustainability [7].

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The treatment strategy for the reduction of pollutants generally depends on its composition and the characteristics of the leachate [8]. Young leachates are basically treated more easily as compared to the old ones. Several technologies which includes coagulation-flocculation, membrane filtration, electro-chemical and advanced oxidation process (AOPs). These methods are very difficult, expensive and generally requires multiple processes, time consuming, and requires skilled personnel to operate [9]. Adsorption is widely reported to be the most effective technique compared to other treatment processes. Adsorption is gaining interest as one of the effective process of advanced wastewater treatment for the removal of recalcitrant organic compounds from landfill leachate [10].

Current research has focused on the utilization of natural materials considered cheap and abundant in the nature for the removal of organic contaminants in leachate [8–10]. Adsorption capacity of media principally depends on the characteristics of materials such as specific surface area, pore size, and its distribution. Composite materials have also been developed from low cost adsorbents to improve adsorptive properties for the reduction of pollutants [11]. However, previous studies on composite materials were limited to the application for several contaminants removal from landfill leachate. Not much attempt has so far been made to obtain a comprehensive leachate treatment in terms of the treatment efficiency of low-cost adsorbents such as micro peat for the reduction of priority parameters from leachate. Hence, it is essential to explore the use of micro peat as an alternative to commercial AC for the reduction of parameters in landfill leachate. In the present study, experiments were carried out using low cost naturally occurring materials which was used to produce composite adsorbent for the reduction of COD and color from landfill leachate. Micro peat is relatively inexpensive abundant material and had been extensively used for the removal of many contaminants from wastewater. Various studies had indicated the capabilities of micro peat as an adsorbent and filter material in leachate treatment processes [12,13]. Micro peat is a highly organic substance derived primarily from plant materials and a representative material of soft soils [14]. Micro peats are those soils that have an organic matter more than 65% or less than 35% of the mineral content and highly porous with a surface area of more than 200 m^2/g [15,16]. The synthesis of adsorbents as composite has better adsorption capacity than the use of single adsorbent [17,18]. The effectiveness of composite adsorbent depends on the compactibility of the starting materials and also the physical, chemical and mechanical stability. This influences the efficiency of the porous adsorbent for the removal of pollutants from the analyte [19,20]. The effectiveness of composite is determined by the adsorption capacity which is influenced by the affinity of the pollutant to the adsorbent. Mesoporous activated carbon is suitable for the adsorption of non-polar solutes due to carbon–oxygen surface groups. Peat is biodegradable and a renewable resource which contains oxygen functional groups such as carboxyl and carbonyl groups. Peat can be used as a partial replacement of commercialized carbon which is known to be expensive. AC-Peat composite has the potential of enhancing the carbon-oxygen surface groups and the efficiency can influence the removal of pollutants from the adsorbate [21,22]. The present study aims to evaluate the efficiency of micro peat activated carbon composite for the removal of COD and color from landfill leachate, in addition to describe the pattern of adsorption of the pollutants using the expression of isotherms and kinetics equations. The experiment is conducted as partial replacement of AC with MP as an alternative media to improve the adsorption properties and the reduction of the treatment cost.

2. Materials and methods

2. 1. Leachate sampling

The sampling location for the raw leachate was at Simpang Renggam landfill site, Johor, Malaysia. This site experiences highly colored and turbid leachate due to the presence of a combination of organic substances and suspended solids [23]. The collection and preservation of samples was achieved in accordance with the Standard Methods for the Examination of Water and Wastewater [24]. Samples were immediately transported to the laboratory in a air tight 10 L plastic container and stored in a cold room at 4ºC prior to use for experimental purposes to avoid the chemical and biological alterations of the leachate characteristics. Chemical analysis was performed within 48 h [24]. All chemicals used for the experimental analysis were of analytical grade.

2.2. Preparation of media

Activated coconut shell carbon was locally purchased at RM4500 per ton. Micro peat sample was collected at Kampung (Kg.) Parit Nipah, Parit Raja, Batu Pahat, Johor, Malaysia. The sampling and preparation were conducted according to the procedure outlined by previous researchers [25,26]. Prior to the experiment, the composition of AC and MP was determined by X-ray fluorescence spectrometry (XRF) (Model Bruker S4 Pioneer). The density of both media was determined conventionally by considering weight/volume of media. AC and MP were sieved to obtain particle size of less than 150 µm. The general properties of media shown in Table 1.

2.3. Batch adsorption experiments

To maximize organic pollutants removal by the adsorbent, batch experiments were conducted at ambient temperature using the optimum conditions of all pertinent factors, such as dose, pH, shaking speed, and contact time. Subsequent adsorption experiments were carried out with only optimized parameters. The optimum conditions for the adsorption batch study taken from the previous study are pH 7, 200 rpm of shaking speed and 120 min of contact time [11]. A similar pH range was reported for COD and color removal in other batch adsorption studies [3,25].

2.4. Optimum mixing ratio

The determination of optimum mix ratio between AC and MP was determined based on previous studies pro-

Table 1
Composition of AC and MP [5]

Component	AC (%)	MP (%)
C	93	23.42
CaO	2.3	2.96
SiO_2	2.3	48.18
Al_2O_3	0.31	9.55
MgO	0.35	2.44
K_2O	0.2	0.26
Fe_2O_3	0.2	4.81
TiO_2	–	0.21
SO_3	0.6	7.67
Na_2O	–	0.32
Others	0.74	–

posed for various quantity of media (by weight) [11,25]. The total weight of the media mixture used was 4.0 g for each conical flask. The sequence of the media mix ratios used were 0:4.0, 0.5:3.5, 1.0:3.0, 1.5:2.5, 2.0:2.0, 2.5:1.5, 3.0:1.0, 3.5:0.5, and 4.0:0 for AC to MP. The batch equilibrium studies were conducted by mixing a fixed amount of media (as mentioned above) with 100 mL of raw leachate sample in a 250 mL conical flask at pH 7, shaking speed 200 rpm and 120 min contact time. The optimum mix ratio between both media was obtained in terms of achievable maximum treatment efficiency of COD and color evaluated according to the following equation [27]:

$$\%R = [(C_o - C_e) / C_o] \times 100 \quad (1)$$

2.5. Adsorption equilibrium

The isotherm experiments were carried out by contacting 4.0 g of the MP-AC media with 100 mL of different leachate concentrations (100–10 degree dilution) in 250 mL conical flask allowing sufficient time, 120 min, for adsorption equilibrium. The amount of adsorbed MP-AC (mg/g) was calculated based on a mass balance equation as given by the following equation [28]:

$$q_e = (C_o - C_e)V / m \quad (2)$$

The data obtained were tested against the linearized forms of Langmuir, Freundlich and Dubinin-Radushkevich (D-R) isotherms.

The Langmuir isotherm describes quantitatively the formation of a monolayer adsorbate on the outer surface of the adsorbent, and after that no further adsorption took place [29]. The linearized form of the Langmuir equation is given in Eq. (3);

$$1/q_e = [1/q_m K_L] 1/C_e + 1/q_m \quad (3)$$

The constants of q_m and K_L were obtained from the slope and intercept of the Langmuir plot of $1/q_e$ vs $1/C_e$ respectively. The essential features of the Langmuir isotherm can be in terms of a dimensionless constant called separation factor (S_F) that explains the favorability of adsorption process which is defined by the following Eq. (4);

$$S_F = 1 / 1 + K_L C_o \quad (4)$$

The value of S_F indicates the adsorption nature to be unfavourable ($S_F > 1$), linear ($S_F = 1$), favourable ($0 < S_F < 1$) or irreversible ($S_F = 0$) [30].

The Freundlich isotherm is an empirical model not limited to monolayer coverage alone (occurs on a heterogeneous adsorbent surface), an adsorption isotherm lacking a plateau but also describe multilayer adsorption [31]. The linearized form of the Freundlich equation is given in Eq. (5);

$$\ln q_e = \ln K_F + 1/n_F \ln C_e \quad (5)$$

The constants of K_F and n_F are obtained from the intercept and slope of the Freundlich plot of $\ln q_e$ vs $\ln C_e$ respectively. The value of n_F satisfies the condition $|1| < n_F < |10|$ and represents that the adsorption process is favourable, otherwise it is unfavourable [32].

The D-R isotherm is a more general model in which assumption is not based on homogenous surface or constant adsorption potential, it gives insight into the biomass porosity as well as the adsorption energy [33]. The value of adsorption energy further provides information as to whether adsorption process is physical or chemical in nature. The linearized form of the D-R equation is given in Eq. (6);

$$\ln q_e = \ln q_s - K_{D\text{-}R}\varepsilon^2 \quad (6)$$

E and ε are expressed by Eqs. (6a) and (6b) respectively;

$$E = \sqrt{[1/2K_{D\text{-}R}} \quad (6a)$$

where $K_{D\text{-}R}$ is denoted as the isotherm constant.

$$\varepsilon = RT\ln[1+1 / C_e] \quad (6b)$$

The constants of q_s and $K_{D\text{-}R}$ obtained from the intercept and slope of the D-R plot of $\ln q_e$ vs ε^2 respectively.

2.6. Adsorption kinetics

The kinetics experiments were carried out by contacting 4.0 g of the MP-AC media with 100 mL of leachate with specific initial concentration in 250 mL conical flask and the working range of contact time between ranges 5–120 min.

Kinetic data were analyzed against the linearized forms of pseudo first-order kinetic model, pseudo second-order kinetic model and intra-particle diffusion equations [3].

The linearized form of the pseudo first-order kinetic model equation is given in Eq. (7);

$$\ln (q_e - q_t) = \ln(q_e) - k_1 t \quad (7)$$

The plot between $\ln (q_e - q_t)$ and t gives, k_1 and q_e values.

Pseudo second-order kinetic model was also applied to analyze kinetic adsorption for liquid solution kinetic adsorption in the following linearized form is given in Eq. (8);

$$t/q_t = 1/k_2q_e^2 + 1/q_e\,(t) \quad (8)$$

The values of constants (k_2 and q_e) can be obtained from the intercept and slope of the linear plot of t/q_t versus t.

Intra-particle diffusion kinetic model determines the rate of parameter using the following equation in Eq. (9);

$$q_t = k_i t^{0.5} + c \quad (9)$$

The plot between q_t and $t^{0.5}$ indicates the theoretical values of k_i of the intra-particle diffusion and the c can be obtained from the slope and intercept in respectively.

2.7. Sample analyses

COD was determined by closed reflux and colorimetric method (5220 D) while color was reported as true color assayed at 455 nm using a HACH/DR6000 spectrophotometer and reported as platinum-cobalt (Pt-Co) method, the unit of color being produced by 1 mg platinum/L in the form of chloroplatinate ion. All method was adapted from the Standard Methods for the Examination of Water and Wastewater [18]. All tests were conducted in triplicates to obtain consistent results at room temperature and the maximum analytical error was found to be less than 5%.

3. Results and discussion

3.1. Characteristics of leachate

Initial investigation revealed that the leachate has high concentration of color, COD, and ammoniacal nitrogen (NH_3-N), with lower value of BOD_5:COD ratio (<0.1). Hence, Simpang Renggam landfill can be categorized as stabilized landfill leachate [4,25]. Stabilized landfill leachate is known to contain refractory organic compound [3,23]. The effectiveness of biological process decreases while physico-chemical processes using adsorption in particular on activated carbon (AC) or other adsorbents may become one of the appropriate options. The leachate characteristics of Simpang Renggam landfill are illustrated in Table 2.

Table 2
Simpang Renggam landfill characteristics

Parameter	Values			Std. Dev.
	Min.	Max.	Ave.	
pH	8.05	8.32	8.19	0.11
SS (mg/L)	143	213	177.22	22.63
NH_3-N (mg/L)	1555	2010	1765.34	190.54
COD (mg/L)	2440	2990	2739.06	225.68
BOD_5 at 20°C (mg/L)	156	329	249.45	61.51
BOD_5/COD	0.06	0.12	0.09	0.02
Fe (mg/L)	6.45	8.94	7.19	0.93
Color (Pt-Co)	4061	4748	4539.56	260.00

3.2. Media mix ratio

The influence of different media mix ratio on percentage removal efficiency of COD and color is illustrated in Fig. 1. Based on the figure, it is apparent that removal efficiency of COD and color increased considerably by increasing media ratio between AC and MP from 0 g:4 g/100 mL to 2.5 g:1.5 g /100 mL while color from 0 g:4 g/100 mL to 2.0 g:2.0 g/100 mL respectively. Thus, the best mix ratio between MP and AC of 1.5:2.5 and 2.0:2.0 are selected as the optimal with percentage removal efficiency of 87% COD and 74% color in respectively achieved. This is believed to occur due to the removal efficiency of COD. The removal of color was observed to decrease with further increment of AC which attained the maximum removal rate of both parameters and therefore the increase of available sorption sites may result in aggregation which can decrease the probability of molecules contacting all available adsorption sites [4,25].

3.3. Adsorption isotherms

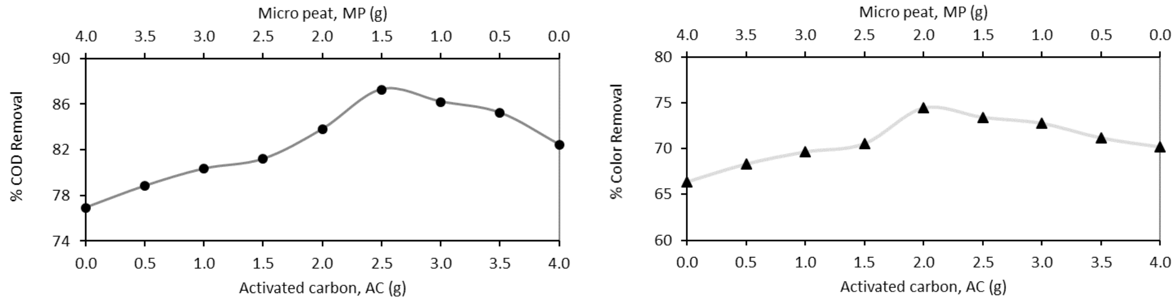
The variations of Langmuir, Freundlich and D-R adsorption isotherms with COD and color onto MP-AC composite media by linear analysis are shown in Fig. 2 while Table 3 summarizes the corresponding isotherm parameters and their correlation coefficients (R^2).

Langmuir isotherm best described the uptake of COD reduction ($R^2 = 0.9205$) and color removal ($R^2 = 0.9007$) onto MP-AC composite media, this suggests that adsorption was not onto a homogeneous distribution of active site but rather occurred on a monolayer surface in organic-MP-AC system. However, multilayer adsorption also played a very important role in the COD reduction ($R^2 = 0.9099$) and color removal ($R^2 = 0.8878$) onto MP-AC composite media. The maximum monolayer adsorption capacity for the uptake of COD and color onto MP-AC composite media was obtained at 23.92 mg/g and 97.09 mg/g, respectively, larger percentage of the adsorbed COD (1.5:2.5) and color (2.0:2.0) parameter was achieved.

The calculated S_F values as different initial leachate concentrations are shown in Fig. 3. It was observed that the value of S_F in the range 0–1 confirms the favourable uptake of the leachate process. Also lower S_F values at higher initial leachate concentrations showed that adsorption was more favourable at higher concentration. Generally, the degree of favourability is related to the irreversibility of the system, giving a qualitative assessment of the MP-AC-organic in terms of COD reduction and color removal interactions. The degrees was observed towards zero which indicated that the completely ideal irreversible case rather than unity better explains the pattern of adsorption [30].

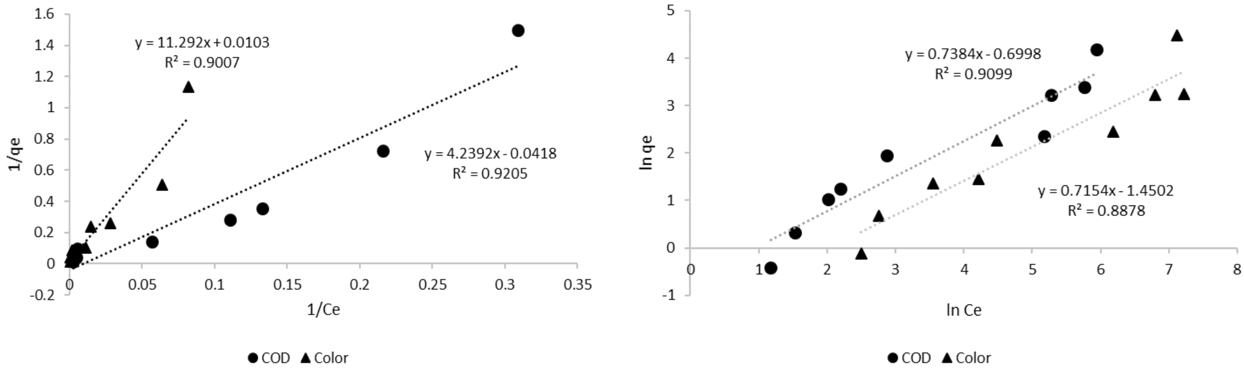
The value of n_F obtained 1.35 for COD and color 1.35 vs 1.40 respectively (Table 3) was greater than 1, indicating that the adsorption process is favourable. If $n_F = |1|$, the adsorption is linear, for $n_F < |1|$, the adsorption is by chemisorption, and for $n_F > |1|$, the adsorption is a favorable physical process [32].

The adsorption energy (E) obtained from D-R isotherm of COD and color were 267.26 kJ/mol and 79.06 kJ/mol (Table 3), respectively suggesting that the uptake of COD

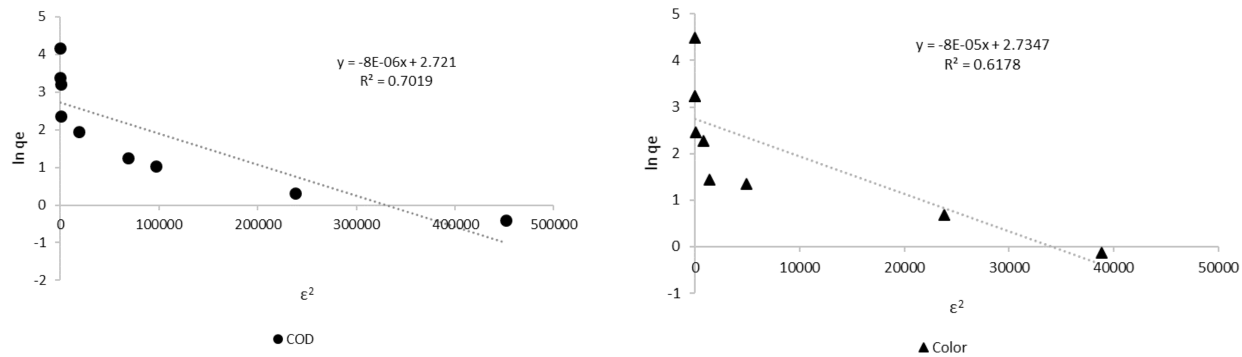


(a) (b)

Fig. 1. Percentage of (a) COD and (b) color removal efficiency against different ratios between MP and AC.



(a) (b)



(c) (d)

Fig. 2. Plots of (a) Langmuir, (b) Freundlich, and (c) D-R for COD and color adsorption using MP-AC composite; $C_{i\,COD}$ = 30–2990 mg/L, $C_{i\,Color}$ = 47–4748 mg/L, 4.0 g composite, < 150 µm, pH 7, 300.15 K, 120 min, 200 rpm.

and color onto MP-AC composite media was by intra-particle diffusion. If the value of E is below 8 kJ mol^{-1} , it indicated a physical adsorption process while if the value of E is between 8–16 kJ mol^{-1}, the process of adsorption is influenced by ion exchange and if E is above 16 kJ mol^{-1} the process is dominated by intra-particle diffusion [33].

Comparing the coefficient of determination values obtained for each of the considered isotherms, the fitting degree follows in order: Langmuir (best fit) > Freundlich > D-R. This trend was also in general agreement with the findings of other researchers [3,29].

3.4. Adsorption kinetics

The applicability of the pseudo first-order, pseudo second-order and intra-particle diffusion kinetic models were tested for the adsorption of COD and color onto MP-AC composite media. The best fit model was selected based on both linear regression coefficient (R^2) and the q_e (calculated) values.

Evidence for this was provided by the linear analysis of the pseudo first-order, pseudo second-order and intra-particle diffusion kinetic models with COD and color onto MP-AC composite media as shown in Fig. 4 while Table 4

Table 3
Langmuir, Freundlich and D-R adsorption isotherm constants

Adsorption isotherm models and parameters		COD	Color
Langmuir	q_m (mg/g)	23.92	97.09
	K_L	9.86×10^{-3}	9.12×10^{-4}
	R^2	0.9205	0.9007
Freundlich	$\lvert n_F \rvert$	1.35	1.40
	K_F	0.4967	0.2345
	R^2	0.9099	0.8878
D-R	K_D (mol²/kJ²)	7.0×10^{-6}	8.0×10^{-5}
	E (kJ/mol)	267.26	79.06
	q_s (mg/g)	12.26	15.41
	R^2	0.7019	0.6178

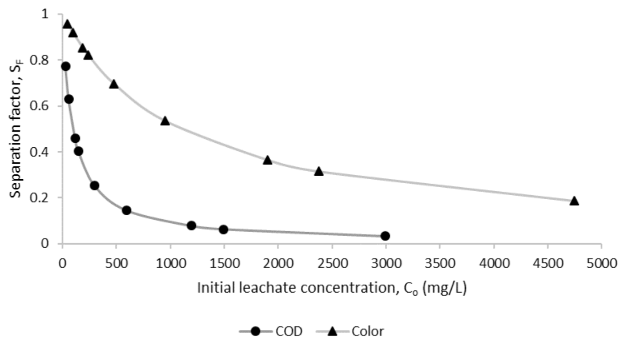


Fig. 3. Plot of separation factor versus initial leachate concentration.

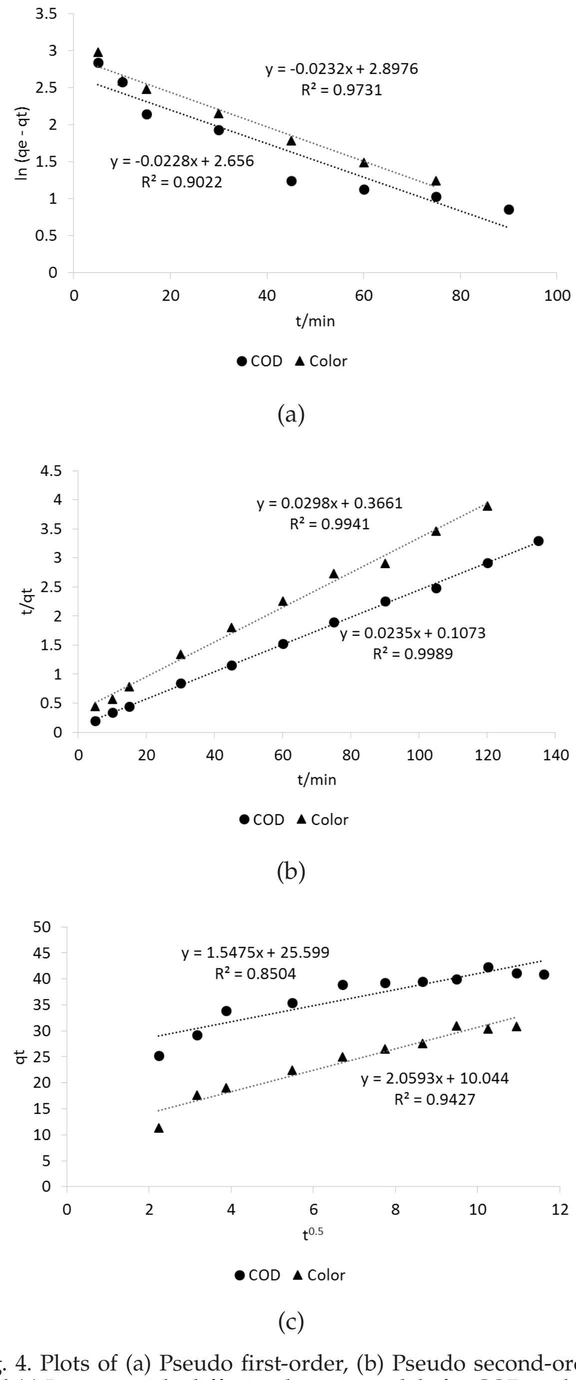


Fig. 4. Plots of (a) Pseudo first-order, (b) Pseudo second-order, and (c) Intra-particle diffusion kinetic models for COD and color adsorption using MP-AC composite; $C_{i\ COD}$ = 2990 mg/L, $C_{i\ Color}$ = 4748 mg/L, 4.0 g composite, < 150 µm, pH 7, 300.15 K, 5–120 min, 200 rpm.

summarizes the corresponding kinetic parameters and their correlation coefficients.

In view of the results obtained, the adsorption of COD and color onto MP-AC composite media is not a first-order kinetic reaction and intra-particle diffusion. However, the correlation coefficient (R^2) for the second-order kinetic model was approximately close to unity for both parameters indicating the applicability of this kinetic equation and the second order nature of the adsorption process of organic constituents onto MP-AC composite media. The theoretical q_{e2} values for organic constituents were also very close to the values deduced from the experimental values. The result suggests that the adsorption kinetics of COD and color were controlled by the chemisorption process that occur at the surfaces and pores of MP-AC composite media. The adsorption process chemically involves monolayer adsorption because of specific bonding between adsorbates and the surface of adsorbent [34]. The results are in agreement with the Langmuir isotherm data of COD and color as previously discussed.

The results are shown in Table 4 corresponds to the external surface uptake and based on these results it might be concluded that intra-particle diffusion was involved in organic constituents adsorption onto MP-AC composite media, but it was not the sole rate determining step and that some other mechanism also play an important role. This is in agreement with similar finding reported by other researchers [3,35].

4. Conclusion

This work was devoted to assess the capability of MP and AC as composite media in order to determine the opti-

Table 4
Kinetic parameters for adsorption rate expressions

Parameter	$q_{e\,exp.}$ (mg/g)	Pseudo first-order			Pseudo second-order			Intra-particle diffusion	
		q_{e1} (mg/g)	k_1 (min^{-1})	R^2	q_{e2} (g/mg min)	k_2 (min^{-1})	R^2	k_i (mg/g min$^{-1/2}$)	R^2
COD	42.32	14.24	0.0228	0.9022	42.55	0.0051	0.9989	1.5475	0.8504
Color	30.97	18.13	0.0232	0.9731	33.56	0.0024	0.9941	2.0593	0.9427

mal amount of mixture ratio for use in the removal of COD and color from a stabilized landfill leachate treatment. It should be noted that treatment with combination of MP and AC for the COD and color gave the best removal efficiency up to 87% (1.5:2.5) and 74% (2.0:2.0) respectively. Among the three adsorption isotherms tested, Langmuir isotherm gave the best fit, followed by the Freundlich isotherm and then the D-R isotherm. The chemisorption was not the sole rate controlling factor. From the values of the apparent energy of adsorption, the adsorption process was found to be a diffusion intraparticle process. The adsorption kinetic study found that the adsorption of both parameters onto MP-AC composite media obeyed the pseudo second-order model (R^2 for COD is 0.9989 and color is 0.9941) respectively. These results reveal that chemical adsorption was the mechanism which was dominant in organic constituents adsorption onto MP-AC composite media. Intra-particle diffusion model indicates that diffusion is not the only rate controlling step since the linearized plot did not pass through the origin. Micro peat has good potential for use as a cost-effective medium to replace activated carbon for organic matter removal at a considerably lower cost. In addition, the use of low-cost media may contribute to the sustainability of the surrounding environment.

Acknowledgements

The authors would like to express their gratitude to the Ministry of Higher Education Malaysia (MyBrain15-MyPhD), Universiti Tun Hussein Onn Malaysia for the financial assistance and also to Universiti Sains Malaysia, Penang, Malaysia for facilities and continuous support.

Symbols

C_o — Initial COD and color concentration in leachate (mg L^{-1})
C_e — Final COD and color concentration in leachate (mg L^{-1})
q_e — The equilibrium adsorption capacity per gram dry weight of the adsorbent (mg g^{-1})
q_m — The maximum monolayer coverage capacity (mg g^{-1})
K_F — Freundlich isotherm constant (mg g^{-1})
q_s — The theoretical monolayer saturation capacity (mg g^{-1})
K_{D-R} — The activity coefficient useful in obtaining the mean sorption energy
E — The mean sorption energy (kJ/mol)
k_1 — The rate constant (min^{-1}) of pseudo-first order model
k_i — The intra-particle (pore) diffusion rate constant (mg/g min$^{-1/2}$)
R — Removal efficiency (%)
V — The volume of the solution (L)
m — The dry weight of adsorbent (g)
K_L — Langmuir isotherm constant (L mg^{-1})
$|n_F|$ — Adsorption intensity
R — The gas constant (J/mol K)
T — The temperature (K)
ε — The Polanyi potential
k_2 — The rate constant of pseudo-second-order model (g/ (min mg))
c — The thickness of the boundary layer

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