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Ammonia Removal Using Organic Acid Modified Activated Carbon From Landfill Leachate

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ABSTRACT

This study was conducted to enhance the ability of activated carbon to remove ammonia by modifying its surface as ion exchanger. The study involved the determination of the capabilities of modified activated carbon to remove ammonia from leachate using fixed bed column. Adsorption capability was determined based on the percentage of ammonia removal. Thomas and Yoon and Nelson models were used to determine the adsorption capacity for both modified and bare activated carbon. Results indicated that modified activated carbon has higher removal ability for ammonia with the maximum removal up to 94.30% compared to bare activated carbon with maximum removal of 64.05%. Based on Yoon and Nelson model, the maximum capacity value, qo was found to be 3.063 mg/g using modified activated carbon which is higher than bare activated carbon with the value of 1.478 mg/g. It can be concluded that the modified activated carbon has a higher ion-exchange capacity for removing ammonia removal compared to bare activated carbon.

1. INTRODUCTION

Landfill is the most widely used method for the disposal of solid waste in Malaysia. Approximately 95% of the total worldwide solid waste disposed in landfills (Kurniawan et al. 2006). Waste that is not properly managed can cause serious threat on human health and problems to the surrounding environment. Production of liquid leachate is among the main issues at landfill sites (Boni et al. 2006). There are several methods of treatment used to treat leachate generated from landfills such as the biological, physical and chemical treatments, combination of physico-chemical treatment such as adsorption, chemical precipitation, membrane filtration, reverse osmosis, ion exchange, air stripping and breakpoint chlorination (Tchobanoglous et al. 2003).

Biological treatment processes are often used to treat young leachate because of its reliability, simplicity and cost effectiveness but less effective for treating matured leachate due to its unstable organic materials and difficult to biodegrade (Robinson et al. 2006). However, physico-chemical methods such as adsorption using activated carbon found to be effective to remove almost 99.99% of organic pollutants including organic materials that are susceptible to biodegradation process (Abu-Zeid et al. 1995). Among the factors contributing to the effectiveness of carbon as adsorbent are a high surface area, high adsorption capacity, microporous structure and reactivity of the surface (Okolo et al., 2000; Robinson et al. 2006). However, an existing treatment using activated carbon unable to remove inorganic materials efficiently. Activated carbon adsorption capacity is not sufficient to adsorb ammonia due to the nature of the surface that is not polar (Park & Kim, 2005) thus lead to weak interaction between bare activated carbon and ammonia (Azhar et al. 2010). Studies have to be done to change the surface of activated carbon to produce an adsorbent that is able to adsorb polar and non-polar materials at the same time. Therefore, this study was conducted to determine the effectiveness for removal of ammonia from leachate using activated carbon surface modified with organic acids found in the leachate.

MATERIALS AND METHOD

Leachate sampling

Sampling for leachate was conducted in Pulau Burung Landfill site located near Byram Reserve Forest (5o24' N, 100o24' E) in Penang. Leachate samples were collected from an active reservoir containing leachate over 5 years, transferred into a 10 L plastic container and stored at 4 °C in the laboratory.

Preparation of modified activated carbon absorbent

In this study, activated carbon with a diameter of about 3.0 mm was used as absorbent. Leachate was passed on to commercial activated carbon until it was saturated to provide the modified activated carbon as ion exchange. The saturated activated carbon was passed on to the regeneration solution produced using sodium chloride (NaCl) 1.0 M with pH adjusted using sodium hydroxide (NaOH). It subsequently rinsed with distilled water and allowed to dry.

Fixed-bed column adsorption

The study was conducted using a column built from polyacrylic with a height of 30 cm and a diameter of 2.5 cm containing 102.71 g activated carbon adsorbent. Samples of leachate were flowed into the column with the help of MASTERFLEX peristaltic pump with a flow rate of 10 ml per minute. The adsorption of leachate was collected at different time intervals of 0 minute, 3 minutes, 5 minutes, 10 minutes, 13 minutes, 17 minutes, 20 minutes, 24 minutes, 29 minutes, 34 minutes, 39 minutes, 44 minutes, 50 minutes and 60 minutes. Leachate was flowed into the column until no further adsorption observed. The concentration of ammonia in the leachate was determined by using Nessler method (USEPA) using a spectrophotometer DR 2700. COD concentration was determined by using the closed reflux colorimetric method using high range digestion reagents (APHA 2005). Whereas the standard method APHA Platinum-Cobalt (Method D1209) was used to determine the concentrations of colour of leachate samples (APHA 2005).

RESULTS AND DISCUSSION

Initial concentration

The initial concentration (before treatment) of ammonia, COD and colour in the leachate were determined and shown in the Table 1.

Table 1: The initial concentration of pollutants in the leachate

Parameter	Concentration
Ammonia	790 mg/L
COD	1676 mg/L
Colour	4420 mg/L PtCo
Organic Acid (total)	130 mg/L

Activated carbon was modified by passing the samples of leachate until saturated. Adsorption of organic acids was obtained at 81.79% for the first 60 minutes with the percentage of removal of 17.44% (Figure 2). A low percentage after 60 minutes of treatment implying minimum organic acids removal observed. This indicates that the surface of activated carbon to be modified has been saturated with organic acids. Organic acids are adsorbed on the surface of activated carbon and suitable for ion exchange because leachate contains a lot of organic matter and carboxylic acids (acetic, propionic and butyric) that are the major organic elements in the leachate from stable landfill (Onal 2006).

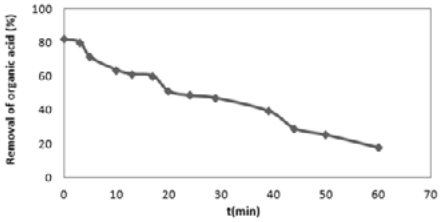


Figure 2: Percentage of organic acid removal by activated carbon

Removal of ammonia

Figure 3 shows the removal of ammonia using bare activated carbon that ranges between 10.13% to 64.05%, while the percentage of ammonia adsorption using modified activated carbon range between 20.04% to 94.30% for the 60 minutes of treatment time (Figure 3). We found that the modified activated carbon is more effective to remove ammonia compared to bare activated carbon. Statistical analysis using Mann-Whitney test showed significant differences in the percentage of ammonia removal using modified activated carbon compared to bare activated carbon with the value of p = 0.0204 (p <0.05).

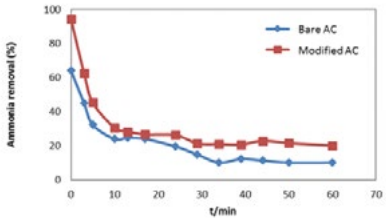


Figure 3: Comparison of ammonia removal between bare activated carbon versus surface modified activated carbon

This is due to the existence of ion exchange surface as a result of the presence of organic acids on the surface of modified activated carbon (Azhar et al. 2012). According to Liu et al. (2007), mechanism of activated carbon ion exchange is due to the presence of functional groups such as carboxylic, on its surface, which has a high affinity and provide surface for adsorption of ionized molecules. The regeneration solution was reacted with an organic acid on the surface of activated carbon to form a carboxylic salt molecule (R-COO-Na +). Regeneration solution with a high pH (pH 12) causes the dissociation of organic acid functional groups on the surface of activated carbon and provides the binding site for the molecule Na + (Chen et al. 2003). This can be explained by the equation below:

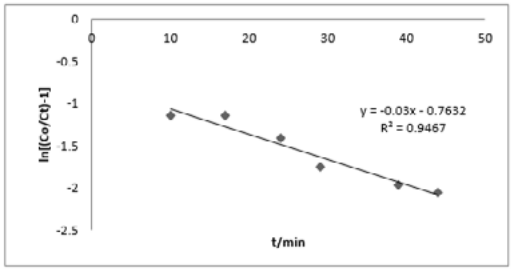
[ADSORBENT]-R-COOH (p) + NaOH (ak)
[ADSORBENT]-R-COO-Na+ (ak) + H2O

Organic acids bind sodium ions with a weak electrostatic bond. This allows the movement and exchange of sodium ions with ammonium ions in the leachate occurs easily. Hydrophilic portion (-COONa) salts of carboxylic undergo ion exchange process with ammonium ions as follows:

[ADSORBENT]-R-COO-Na+ (p) + NH4+ (ak)
[ADSORBENT]-R-COONH4 (p) + Na+

For bare activated carbon, the bond that exists between the non polar activated carbon and polar ammonium ion clusters is very weak thus resulting in low ammonia adsorption by bare activated carbon. This explains why the surface modified activated carbon is more effective in removing ammonia compared to bare activated carbon. The maximum adsorption capacity of ammonia, qo was determined using the Thomas model (1944) and Yoon and Nelson model (1974). Figure 4 shows the linear graph of ln [(Co/C)-1] versus the time interval (minutes) that were plotted based on the Thomas model equation:

where:
kT (mL/min mg) = constant Thomas
qo (mg/g) = maximum capacity of material
mc (g) = mass of absorbent
Q (mL/min) = rate of liquid flow into column
Co (mg/L) = initial concentration
t (min) = time



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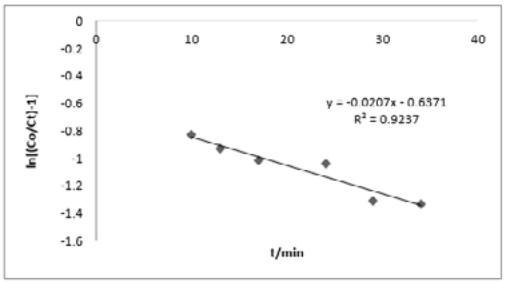


(b)

Figure 4: Linear plot of Thomas model for ammonia absorption using (a) bare activated carbon (b) modified activated carbon

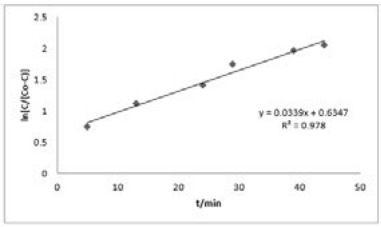
Linear plot of ln [(C0/Ct)-1] against treatment time (t) was used to determine the value of KT and qo based on the intercept and slope of the plot (Table 2). Based on the Thomas model, the value of qo bare activated carbon and modified activated carbon are 1,957 mg/g and 2.450 mg/g, respectively. This implies that the modified activated carbon has a higher qo and more effective for removing ammonia compared to bare activated carbon.

Table 2: Value of R^2, constant kT and q_o for Thomas model

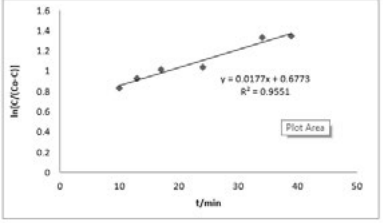
Absorbent	kT (mL/mg/min)	R^2	Maximum adsorption capacity, qo (mg/g)
Bare activated carbon	$3.8x10^{-5}$	0.946	1.957
Modified activated carbon	$2.53x10^{-5}$	0.923	2.450

Figure 5 shows the linear graph of ln [(Ct/C0-Ct)] versus the time interval (minutes) that were plotted based on the Yoon and Nelson model equation:

where:
kYN (1/min) = constant Yoon and Nelson
t0.5 (min) = time (50% of material absorbed)
C0 = initial concentration (inlet)



(a)



(b)

Figure 5: Linear plot of Yoon and Nelson model for ammonia adsorption using
(a) bare activated carbon (b) modified activated carbon

Linear plot of ln [Ct / (C0-Ct)] against treatment time (t) was used to determine the value of kYN based on the intercept and slope of the plot (Table 3). Based on the Yoon and Nelson model, the value of qoYN bare activated carbon and modified activated carbon are 1.478 mg/g and 3,063 mg/g., respectively

Table 3: Value of R^2, constant kT and q_o for Yoon and Nelson model

Adsorbent	Constant k_{YN} (mL/mg/min)	R^2	$t_{0.5}$ (min)	Maximum adsorption capacity , qoYN (mg/g)
New activated carbon	0.033	0.978	19.21	1.478
Regenerated activated carbon	0.017	0.955	39.82	3.063

CONCLUSION

In conclusion, we found that the surface modified activated carbon was capable of removing ammonia from leachate more effectively. The surface modified activated carbon as an ion exchanger showed higher percentage of removal and maximum adsorption capacity (qo) compared to the bare activated carbon. However, the range of removal percentage and the maximum adsorption capacity of COD and colour by both adsorbents showed that bare activated carbon is more effective than modified activated carbon.

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