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Ammoniacal nitrogen and COD removal from semi-aerobic landfill leachate using carbon-mineral composite adsorbent

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Abstract

Chemical oxygen demand (COD) and ammoniacal nitrogen were the problematic parameters in landfill leachate treatment. Combination of activated carbon and zeolite as filter medium may reduce this problem. This study was conducted to find treatment alternative by combining the low cost adsorbent such as limestone and rice husk carbon waste and ordinary adsorbent media, activated carbon and zeolite as a single media. All the adsorption media was crushed and sieved to a particle size of 150µm. The optimum ratio was predicted by mean of a batch equilibrium experiments. Ordinary Portland Cement (OPC) was used as a binder at 30 percent by weight. Activated carbon and rice husk carbon was grouped as a hydrophobic media where the optimum ratio was 1:1. Zeolite and limestone was in hydrophilic media group which the best ratio was 3:1. The ratio for hydrophilic and hydrophobic media had been chosen as 7:1 accordingly to adsorption behavior of ammoniacal nitrogen and organic constituents (COD) to the media. The optimum conditions for adsorption batch study were found at pH 7, 200rpm in shaking speed and 90 minutes of contact time. The results showed that the equilibrium data were fitted and favorable adsorption by both of the Langmuir and Freundlich isotherms while Langmuir isotherms was slightly better fitted for ammoniacal nitrogen and Freundlich was good for COD removal in term of regression coefficients (R^2). Langmuir adsorption capacities (Q) for ammonia and COD were 43.47mg/g and 256.41mg/g respectively while Freundlich (K_F) were 0.00135mg/g and 0.03891mg/g respectively.

Keywords: Composite adsorbent, leachate treatment, physico-chemical treatment, semi-aerobic leachate

Introduction

Leachate is a liquid formed primarily by the percolation of precipitation water through the open landfill or through the cap of the completed site (Senior, 1995). Leachates may contain large amounts of organic contaminants which can be measured as chemical oxygen demand (COD) and biological oxygen demand (BOD), ammonia, halogenated hydrocarbons suspended solid, significant concentration of heavy metals and inorganic salts (Trebouet et al., 2001; Bagchi, 1990). If not treated and safely disposed, landfill leachate could be a potential source of surface and ground water contamination, as it may percolate through soils and subsoils, causing pollution to receiving waters (Tatsi, et.al. 2003). Most of the landfills do not have leachate treatment facilities. Leachate from this improperly designed landfill may pollute our environment, especially the surface and groundwater. The two major problem of leachate is organic contaminant and ammoniacal nitrogen. Biological treatment of landfill leachates have been shown to be very effective in removing organic matter in early stages (Berruetta & CastrillÓn, 1992) when BOD/COD ratio of the leachate has a high value. This ratio decreases with the age of the landfill (Rodriguez et al., 2000) and the process is less effective with time (Mendez et al., 1989), due to the major present of refractory organic matter.

In recent years, due to cost-effectiveness, the great interest has been focused on the preparation of low cost adsorbents as a substitute or reduces activated carbons consumptions. The development of modern technology requires a continuous search for new adsorbents. Carbon–mineral composite adsorbents can be considered as a new type of adsorbent, which may have structures and adsorption properties different from the individual components (Leboda, 1992 & 1993). In the literature mechanical mixing of carbon and mineral adsorbents, addition of carbon particles to the mineral sol before gelation prepares carbon mineral composite adsorbents, and carbonization of organic substances previously bonded to the mineral adsorbent (Leboda, 1992).

Composite materials had been developed for many purposes such as improve adsorptive properties or to produce low cost adsorbent (Hadjar et. al., 2004). It is well known that activated carbons are the most effective adsorbents for removal of organic pollutants from aqueous or gaseous phase. Therefore this kind of adsorbent is widely applied as a commercial adsorbent in the purification of water and air (Leboda, 1992 & 1993). In recent years, the great interest has been focused on the preparation of low cost adsorbents as a substitute or reduces activated carbons consumptions. Clays are potential materials. Modification of clays for the adsorption purpose has been reported by a number of investigators (Shah et. al, 2000).

The results showed that adsorption effect of organic matters by activated carbon was over than that by zeolite. Zeolite could cut ammonia nitrogen peak value of influent, while activated carbon could remove ammonia nitrogen steadily. Zeolite and activated carbon composite process can efficiently remove ammonia nitrogen and organic matters in micro-pollutant raw water (Deng et. al., 2004). Gao et. al. (2005) found new composite materials of zeolite–carbon (Z–C), which combines the excellent properties of zeolites and carbon. The surface of zeolite is hydrophilic with regular aligned molecular level pores and cationic exchange ability, which makes it a good adsorbent for metallic ions and catalyst (Ono & Yashima, 2000). On the other hand, the surface of carbon is hydrophobic with pores size in the nanometer range or above; hence it is more suitable for the adsorption of organic substances (Okolo et. al, 2000). The resulting Z–C composites with controlled zeolite phase and varied carbon content, therefore, are prominent candidates for the removal of heavy metal ions and organic pollutants, for the purification of exhaust gases, as building materials with heat-insulating and humidity-controlling capacity, and as electromagnetic wave absorbers.

The usages of new composite materials for leachate treatment were the focus of this study. Leachate sample were collected from Pulau Burung Landfill site, Penang, Malaysia. Previous study indicated that COD and ammoniacal nitrogen in this Landfill site was up to 2580mg/l and 1030mg/l respectively and BOD/COD ratio as low as 0.02-0.06 (Aziz et al., 2007). In this study besides combination of activated carbon and zeolite, the low cost adsorbent such as rice husk carbon (RHC) and limestone has been use to replace activated carbon and zeolite partially. In this research we aims to develop a single composite adsorption material based on activated carbon, zeolite and low cost adsorption materials such as limestone and RHC. To achieve this objective we have to determine the optimum mixture for the adsorption materials above based on aggregate organic contaminant (COD) and ammoniacal nitrogen removal.

Materials and Methods

Adsorbent Materials

Limestone chips used in the study were purchased from marble factory, which cost about RM40 per ton. Activated carbon and zeolite were commercially available for at a cost of RM2-4 per kg. The density of media was determined conventionally, i.e., weight/volume of media. Rice husk carbon (RHC) waste was obtained from the rice mill factory at Nibong Tebal. All the media were ground to give particle size less than 150µm using ceramic ball mill. Adsorption materials were classified as main adsorbent-low cost adsorbent and hydrophobic-hydrophilic adsorbent as mentioned in Table 1.

Leachate sample will be collected from Pulau Burung Landfill Site (PBLS); which is situated within Byram Forest Reserve at 5° 24' N, 100 ° 24'E in Penang, Malaysia. Total area of the landfill is 23.7 ha and it is equipped with a leachate collection pond but without other treatments. This site a natural marine clay liner. PBLS has a semi-aerobic system and it is one of the only three sites of its kind found in Malaysia. PBSL has been developed semi-aerobically into a sanitary landfill Level II by establishing a controlled tipping technique in 1991. It will be further upgraded to a sanitary landfill Level III employing controlled tipping with leachate recirculation in 2001. This site receives 1,500 tons of solid will bate daily (Aziz, et. al, 2007).

Samples were collected from the active detention pond with leachate age of less than 5 years and filled in 30-L plastic container, transported to the laboratory and stored at 4°C. Chemical analysis was performed during the following two days, according to Standard Methods for the Examination of Water & Wastewater (1992). All chemicals used for the analytical determinations were of analytical grade.

Table 1 Adsorption materials.

Type of Adsorbent	Main adsorbent	Low cost adsorbent
Hydrophobic	Activated carbon	Rice Husk Carbon (RHC)
Hydrophilic	Zeolite	Limestone

Ordinary Portland cement has been chosen as a binder to fix all the adsorbent together as a single media. Amount of OPC were determined by attrition measurement, modification of procedure by Toles, et. al. (2000). Ten grams of composite media particles (1.18 mm - 2.36 mm diameter) was placed in 100 ml of raw leachate. The mixture was stirred at maximum speed (350 rpm) for five hours at room temperature. The mixture was filtered through a 1.18 mm sieve and washed with distilled water. The media not passing the sieve was quantitatively transferred to a pre-weighed glass watch. The samples were kept at 110 ^{0}C for two hours, allowed to cool to room temperature and the final weight was obtained. The percent attrition was the percent ratio of difference between initial and final weights of media to initial weight. This method was modified to suit the real conditions of this study. In this study, the amount of binder has to be adequate to produce strong enough media that could not be broken during batch study especially in particle size effect experiment. The aim of this experiment was to get the minimum OPC amount that gives minimum attrition percentage.

Optimum Ratio

The determinations of optimum ratio between hydrophobic and hydrophilic media were determined based on the adsorption properties towards ammoniacal nitrogen and COD, the major contaminant in leachate. Batch adsorption study were performs at pH7 and at 5 hours contact time to investigate the adsorption properties that give the optimum ratio. The ratio that gives maximum removal of the both contaminants is considered as the optimum ratio.

Table 2

A. Hydrophobic, activated carbon - rice husk carbon;

AC/g	0.0	0.5	1.0	1.5	2.0	2.5	3.0	3.5	4.0
RHC/g	4.0	3.5	3.0	2.5	2.0	1.5	1.0	0.5	0.0

AC – Activated carbon, RHC – Rice husk carbon

B. Hydrophilic, zeolite - limestone;

Zeolite/g	0	5	10	15	20	25	30	35	40
Limestone/g	40	35	30	25	20	15	10	5	0

C. Combination hydrophobic-hydrophilic

hydrophobic /g	0	1	2	3	4	5	6	7	8
hydrophilic /g	8	7	6	5	4	3	2	1	0

Steps in optimum ratio determination

A. Optimum ratio for hydrophobic media - Activated carbon (AC) to rice husk carbon (RHC). Main hydrophobic media activated carbon will be replaced partially by low cost adsorbent RHC.
B. Optimum ratio for hydrophilic media - Zeolite (Z) to limestone (L). Main hydrophilic media zeolite will be replaced partially by low cost adsorbent limestone.
C. Optimum ratio for combination hydrophobic-hydrophilic media - ratio A to B.

Batch Adsorption Experiment

Optimum conditions of batch study were determined using 5 g of media and 100 ml raw leachate or 50g/L of media concentration. To maximize ammoniacal-nitrogen and aggregate organic (COD) removal by the adsorbent, batch experiments were conducted at ambient temperature using the optimum conditions of all pertinent factors such as dose, pH, agitation speed and contact time. Adsorption isotherm tests were also carried out in the reaction mixture consisting of 50g/L of adsorbent and 100ml of leachate solution with varying leachate concentrations using distilled water.

Ammoniacal-nitrogen and COD analysis

Ammoniacal-nitrogen was determined using Orion Ammonium ion selective electrode. The sample needs to be acidified to ensure that the ammonium ion remains in solution. The ammoniacal-nitrogen analyses were cross-checked with Nesslerisation and titration technique on several samples (Jorgensen and Weatherley, 2002). COD was determined using HACH reactor digestion method for high range determination (Jirka and Carter, 1975).
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Results and Discussion

Media Preparation

The determinations of adsorption media composition were based on adsorption behavior of ammoniacal nitrogen and COD. The optimum composition of hydrophobic adsorbent materials such as activated carbon and rice husk carbon had been determined by varying the ratio of the both materials. The optimum ratio for activated carbon and rice husk carbon was 1:1 where the removal percentage of NH_3 and COD remained constant. The optimum ratio of hydrophilic materials, zeolite:limestone was 3:1 with removal for ammonia was up to 83 percent while for hydrophobic materials, AC:RHC was 1:1 with COD removal was 80 percent as showed in Figure 1 and 2. The best ratio for hydrophobic-hydrophilic adsorbent materials was 1:7 as compromised ammonia and COD removal (Figure 3). Ammonia has been chosen to test hydrophilic media due to its properties that very soluble in aqueous solution whether in ammonia or ammonium forms. Zeolite was a main material for hydrophilic media in this study is an ion exchanger having a high affinity to ammonium ions (McVeigh, 1999; Woods, 1997; Semmens, 1981; Ames, 1967). Almost organic contaminants in wastewater were hydrophobic and could be remove effectively by activated carbon adsorption.

Ordinary Portland cement (OPC) has been chosen as a binder due to its compatibility to adsorption media especially limestone, zeolite and rice husk carbon. At the same time OPC could be an adsorption media. Previous study had indicated that hardened paste of Portland cement has been used as a low cost adsorbent for the removal of arsenic from water environment (Kundu et. al., 2004). Phosphate ions have been removed from aqueous solution by fly ash, slag, ordinary Portland cement and related cement blends (Agyei et al., 2002). Figure 4 apparently showed that the optimum percentage of OPC binder was 30 percent. Finally the composite adsorbent was prepared and their physical and chemical properties were listed in Table 2.

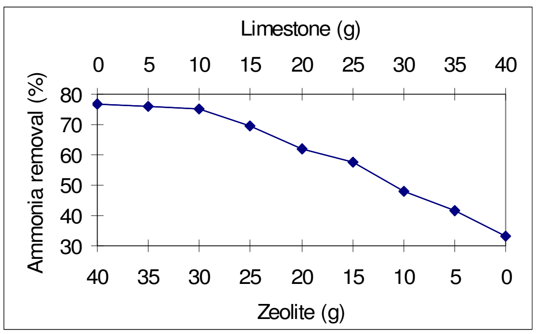

Figure 1 Optimum ratio of zeolite-limestone for hydrophilic media preparation at pH 7 and 200 rpm shaking speed.

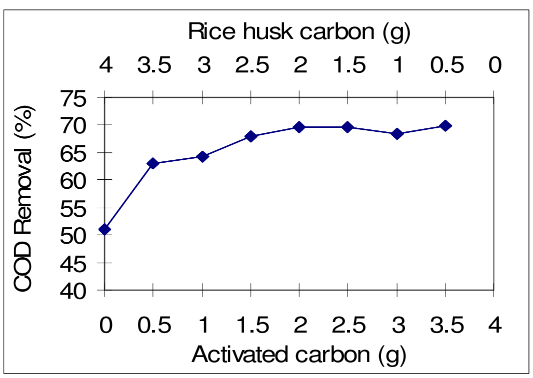

Figure 2 Optimum ratio of activated carbon - rice husk carbon (Table 2A) for hydrophobic media preparation at pH 7 and 200 rpm shaking speed.

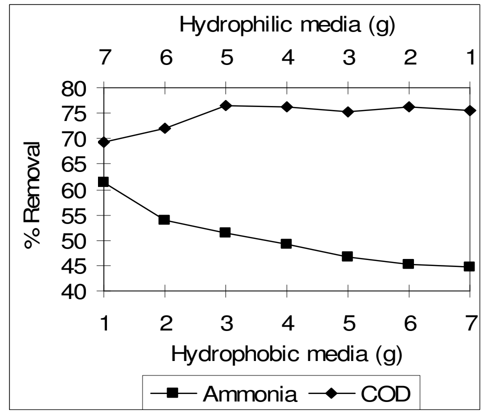

Figure 3 The optimum ratio of hydrophobic-hydrophilic adsorbent (Table 2C) at pH 7, and 200 rpm shaking speed.

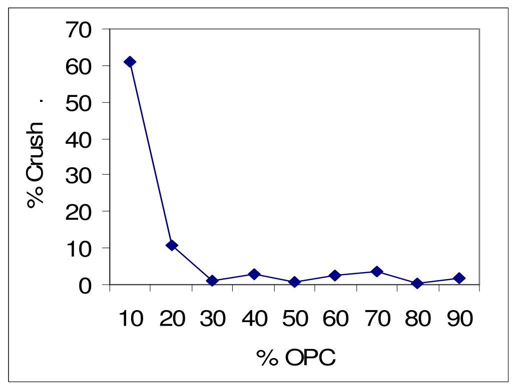

Figure 4 Optimization amount of binder, percentage of ordinary Portland cement (OPC) in composite adsorbent media at pH 7 and 350 shaking speed.

Table 2 Physico-chemical characteristics of composite adsorbent.

Composite Adsorbent Properties	
Specific gravity (g/cm^3)	2.80
BET Surface area (m^2/g)	60.94
Porosity (%)	55.76
Water absorption (%)	52.48
Methylene blue number (mg/g)	6.33
Iodine number (mg/g)	16.92
Cation exchange capacity ,CEC (meq/g media)	0.9204

Leachate charactericties from semi-aerobic landfill site (Pulau Burung, Penang, Malaysia) were listed in Table 3. According to pH (>5) and BOD_5-COD ratio (<0.1), the leachate could be classified as a stabilized leachate (Alvarez-Vazkuez et al., 2004).

Table 3 Semi-aerobic landfill leachate characteristics (April 2006 – Mei 2007)

	Range	Average
pH	8.09-8.66	8.29
Suspended solids (mg/L)	124 – 190	165.57
Ammonia (mg/L)	1010 – 2740	1890.95
COD (mg/L)	1478 – 3540	2338.29
BOD_5 (mg/L)	160 – 345	227.58
BOD_5/COD (mg/L)	0.06 – 0.15	0.09
Fe (mg/L)	2.94-7.30	5.41
Color (mg/L)	3773-5100	4527.36

Adsorption isotherm

Langmuir adsorption model

This adsorption model is based on the assumption that maximum adsorption correspond to a saturated monolayer of solute molecules on the adsorbent surface. The saturated monolayer curve can be represented by the expression;

$$q_e = \frac{QbC_e}{(1+bC_e)} \quad (2)$$

The Langmuir equation can be described by the linearized form

$$\frac{1}{q_e} = \frac{1}{Q} + \frac{1}{QbC_e} \quad (3)$$

where Q and b are Langmuir constant determined from the slope and intercept of the plot, indicative of maximum adsorption capacity (mg/g) and energy of adsorption respectively. The remaining concentration of adsorbate shows by Ce, while qe is the amount adsorbate at equilibrium (mg/g) (Kadirvelu et al., 2001)

The single-solute sorption of ammonia and COD by composite media was plotted by varying real leachate concentration using distilled water (dilution factor from 1.43 to 20) shows in Fig.5 and 6. The concentration of ammonia and COD was determined before performing isotherm adsorption study. The adsorption capacity Q and energy of adsorption b were determined from the slope and intercept of the plot and are represented in Table 4.

According to Kadirvelu et al. (2001), the essential characteristics of Langmuir isotherm can be explained in terms of a dimensionless constant separation factor (R_L), defined by;

$$R_L = \frac{1}{(1+bC_o)} \quad (4)$$

Where b is the Langmuir constant and C_0 is the initial concentration of adsorbates. The value of R_L indicated the type of Langmuir isotherm to be irreversible (R_L = 0), favorable (0 < R_L < 1), linear (R_L = 1), or unfavorable (R_L > 1). In the present study, the values of R_L (Table 5) were observed to be in range of 0-1, indicating that the adsorption of ammonia and aggregate organic constituent (COD) were favorable for this study.

Table 5 Dimensionless constant separation factor, R_L for selected initial concentration for ammonia and COD.

Adsorbates	B (L/mg)	Initial concentration (mg/L)	R_L
Ammonia	3.06×10^{-4}	1994	0.7793
		1395	0.8346
		997	0.8760
		598.2	0.9217
COD	3.641×10^{-3}	2040	0.1187
		1428	0.1613
		1020	0.2121
		204	0.5738

Freundlich isotherm model

The Freundlich isotherm (Chiou, M.S. and Li, H.Y., 2002; Choy Keith et al., 1999) is a special case for heterogeneous surface energy in which the energy in the Langmuir equation varies as a function of surface coverage strictly due to variation of the sorption. The Freundlich equation is given as;

$$q_e = K_F C_e{}^{1/n} \quad (5)$$

where K_F is roughly an indicator of the adsorption capacity and 1/n of the adsorption intensity.

Table 4 Langmuir and Freundlich isotherm constants for adsorption of ammonia and COD onto composite adsorbent.

Adsorbates	Langmuir				Freundlich		
	Q(mg/g)	b(ml/mg)	R^2		K_F(mg/g)	n	R^2
Ammonia	43.47	1.1484×10^{-4}	0.9924		0.00135	0.8137	0.9796
COD	256.41	9.902×10^{-5}	0.905		0.03891	1.08613	0.9502

A linear form of the Freundlich expression will yield the constants K_F and 1/n.

$$\log q_e = \log K_F + \frac{1}{n}\log C_e \qquad (6)$$

Therefore, K_F and 1/n can be determined from the linear plot of log qe versus log Ce. The values are presented in Table 2 and the Freundlich equation isotherms are shown in Fig. 9. The magnitude of the exponent 1/n gives an indication of the favorability of adsorption. Values of n>1 obtained represent favorable adsorption (Chiou, and Li, 2002).

Conclusions

The composite adsorption media had been produced using activated carbon, zeolite and low cost media such as limestone and RHC. The optimum ratio for AC:RHC was 1:1 while zeolite:limestone was 3:1. Overall ratio for (AC-RHC):(zeolite:limestone) was 1:7. The optimum conditions for adsorption batch study were found at pH 7, 200rpm shaking speed and 90 minutes contact time. Both Langmuir and Freundlich isotherm study indicated that this media showed favorable adsorption for ammonia nitrogen COD removals in leachate. Langmuir adsorption capacities (Q) for ammonia and COD were 43.47mg/g and 256.41mg/g respectively while Freundlich (KF) were 0.00135mg/g and 0.03891mg/g respectively. According to the regression value, ammonia adsorption was better fitted to Langmuir model while COD was better using Freundlich model.

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