OPEN ACCESS

Water ISSN 2073-4441 www.mdpi.com/journal/water

Article

The Effectiveness of Silica Sand in Semi-Aerobic Stabilized Landfill Leachate Treatment

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Received: 10 October 2010; in revised form: 1 November 2010 / Accepted: 25 November 2010 / Published: 6 December 2010

Abstract: This study examines the suitability of natural silica sand as a low-cost adsorbent for the removal of ammoniacal nitrogen and heavy metals, particularly iron and zinc, from semi-aerobic stabilized landfill leachate. Leachate samples were collected from the Pulau Burung landfill site (PBLS) in Penang, Malaysia. The above-mentioned contaminants are highlighted in this study because of their unsafe concentrations at PBLS. The effects of shaking time, settling time, and silica sand dosage on the study parameter removal efficiencies were investigated to predict the performance of the process. The adsorptions of ammoniacal nitrogen, iron, and zinc were judiciously described by Langmuir and Freundlich isotherm models. The optimum removal efficiencies of ammoniacal nitrogen, iron, and 39.2%, respectively, with a shaking time of 90 min, a settling time of 60 min, and with a dosage of 60 g (0.5 kg/L) of silica sand. Based on the coefficient of determination (R^2) values obtained from Langmuir and Freundlich isotherm models, ammoniacal nitrogen, iron, and zinc determination were better fitted to the Freundlich model.

Keywords: semi-aerobic landfill; stabilized leachate treatment; adsorption; silica sand

1. Introduction

Landfill is the technique employed most worldwide for the disposal of municipal solid waste (MSW) in developing countries such as Malaysia [1-3]. After being placed in landfills, solid waste habitually decomposes through a series of combined physicochemical and biological processes. Accordingly, the decomposition of waste in combination with percolating precipitation results in the creation of an extremely polluted liquid called landfill leachate. As stated by Kurniawan and Lo [4], one metric ton of landfill MSW would generate 0.2 m³ of leachate; however, the qualities and the quantities of landfill leachate are influenced by the moisture content, site hydrology, landfill age, climatic conditions, and degree of waste stabilization [5].

Landfill leachate contains high amounts of organic compounds, ammonia, and heavy metals [6]. Thus, the generation of landfill leachate creates the potential for long-term impact on the surrounding environment. The potentiality of leachate to eventually find its way into ground/surface water creates serious hazards to public health and ecosystems. Therefore, leachate treatment remains a salient anxiety and a remarkable concern worldwide [7,8]. The leachate composition from different sanitary landfills, as mentioned by Renou et al. [9], displays wide variation. Chemical oxygen demand (COD) concentrations can vary from 100 to 70,900 mgL⁻¹, resulting in severe toxicity in many cases. The BOD₅ (Biochemical Oxygen Demand)/COD ratio (from 0.70 to 0.04) could quickly decrease as the landfill site ages [10], showing the low efficiency of biological treatment. In addition, landfill leachate contains high amounts of ammonia that can reach up to 13,000 mgL⁻¹. Ammoniacal nitrogen has been classified as the primary source of acute toxicity when its concentration is higher than 100 mg/L, as proven in [11]. The highly concentrated pollution of the leachate makes the validity of biological treatment insufficient and impermanent, particularly for stabilized leachate [9,12]. Typically, the age of landfill is a main decisive factor for the selection of an appropriate treatment technique for landfill leachate. The presence of high levels of BOD₅ in young leachate makes it suitable for biological treatment. This method is known to be reliable, simple, and cost-effective [13]. In contrast, physicalchemical methods, which are not preferential for young leachate treatment, have been found to be suitable for the removal of refractory substances from old "stabilized" leachate [9,12].

In most countries, environmental rules and regulations currently enforced by regulatory agencies concerning the observation and control of pollutants resulting from leachate waste streams are becoming more rigid. Imposing such rules inevitably affects the design, planning, and operation of the municipal landfills [14]. This has encouraged growing research awareness focused towards establishing an evidently important, distinctive, consistent, and durable treatment for heavily polluted leachate. Lately, an extensive variety of scientific studies, widely covering collection, storage, and suitable treatment of highly contaminated landfill leachate, has been conducted [9]. In light of the literature, adsorption [1,15], air stripping [16], membrane filtration [17], coagulation, flocculation [7], ion exchange [3,18], chemical and electrochemical oxidation methods [19-20], and chemical precipitation [21] are the major common physicochemical treatment methods for stabilized landfill leachate treatment. Among all the above-mentioned methods, adsorption can be classified as the most competent and promising fundamental approach in wastewater treatment. It is known as a surface phenomenon in which a multi-component fluid (gas or liquid) mixture is attracted to the surface of a solid adsorbent, shaping attachments via physical or chemical bonds [8,22].

In view of the above matter, adsorption via activated carbon has gained prominence in the removal of an indispensable quantity of organic substances, typically measured as COD, from the stabilized leachate. Activated carbon is well known as a typical and effective medium that can successfully remove organic substances from landfill leachate. However, the presence of a considerable amount of inorganic substances (indicated by ammoniacal nitrogen), which is difficult to effectively remove by activated carbon, has necessitated investigation into the performance efficiency of numerous low-cost available natural materials, such as zeolite and limestone, as adsorbents [1,15]. In the present study, the treatability of ammoniacal nitrogen, iron, and zinc from semi-aerobic stabilized landfill leachate via low-cost natural silica sand is investigated and documented. In addition, the equilibrium isotherms in this study are investigated and illustrated with Langmuir and Freundlich isotherms. This experiment is the first of its kind for semi-aerobic stabilized landfill leachate. The results will be thoroughly utilized as a base to investigate cheaper unconventional adsorbents for such landfills.

2. Materials and Methods

2.1. Landfill Site Characteristics

The present study focuses on the leachate generated from the Pulau Burung landfill site (PBLS). PBLS is located on the northwest coast of Peninsular Malaysia, which has a tropical climate (within the Byram Forest Reserve at 5° 24' N Latitude, 100° 24' E Longitude), approximately 20 km southeast of Penang Island. The total area of the landfill is 63.4 ha; nevertheless, only 33 ha are currently operational, receiving 2,200 tons of solid waste daily. This site was developed as a semi-aerobic sanitary landfill Level II by establishing a controlled tipping technique in 1991. In 2001, PBLS was upgraded to a Level III sanitary landfill by employing controlled tipping with leachate recirculation. In Malaysia, the level of improvement of sanitary landfill system can be achieved in four stages [23]:

Level I: Controlled tipping Level II: Sanitary landfill with a bund (embankment) and daily soil covering Level III: Sanitary landfill with a leachate recirculation system Level IV: Sanitary landfill with leachate treatment facilities

2.2. Sampling

Leachate samples were collected manually (10 L per sample) monthly from the aeration pond at PBLS from January to April 2009. In accordance with the Standard Method of Water and Wastewater Examination, the samples were immediately transported to the USM Environmental Engineering Laboratory. The samples were preserved in a cold room at 4 °C prior to experimental use in order to minimize biological and chemical reactions. All chemical analyses for leachate characterization were carried out within the following 24 h. The samples were analyzed for ammoniacal nitrogen, iron, and zinc according to Standard Methods for the Examination of Water and Wastewater [24].

2.3. Silica Sand Properties

Natural raw silica sand was taken from the former mining site in Serdang, Kedah, Malaysia. The raw silica sand was washed repeatedly to remove impurities, roots, leaves, and soils, and then air-dried

under sunlight. Subsequently, it was sieved to 0.6–1.18 mm particle size. After drying, the chemical composition of the silica sand was determined by x-ray fluorescence (Rigaku RIX3000), as illustrated in Table 1. The physical properties of the silica sand applied in this study are provided in Table 2. The density of silica sand was measured by gas pyrometer test (Micromeritics AccuPyc 1330). Brunauer-Emmett-Teller method using the Micromeritics FlowSorb II 2300 was used to determine the specific surface area (total area available for adsorption).

Composition	Results (%)	Composition	Results (%)
SiO ₂	92.214	Fe ₂ O ₃	0.236
Al_2O_3	5.848	SO_3	0.013
K ₂ O	1.382	Rb ₂ O	0.010
MgO	0.088	ZrO_2	0.007
Na ₂ O	0.071	NiO	0.004
TiO ₂	0.051	ZnO	0.002
CaO	0.040	CuO	0.001
P_2O_5	0.032		

Table 1. Chemical composition of silica sand.

Type of test	Result
Surface area, (m^2/g)	0.33
Density, (kg/m ³)	2510
Void ratio, (%)	50
Particle size, (mm)	0.60-1.18

Table 2. Physical characteristics of silica sand.

2.4. Experimental Conditions

In this study, a series of batch experiments were carried out in order to determine the optimum experimental conditions to achieve the maximum removal efficiency of NH_3 -N, iron and zinc. All experiments were conducted by shaking 120 mL of raw leachate with a specific amount of silica sand in a 250 mL conical flask at a shaking speed of 350 rpm using an orbital shaker (model PROTECH 720, Malaysia). The influence of shaking time, settling time and silica sand dosage on the removal parameters was examined. The influence of the shaking time was initially evaluated by running the experiments for many consecutive times for different durations (30, 60, 90, and 120 min), with a shaking speed of 350 rpm, settling time of 2 h and silica sand dosage of 40 g (333 g/L). The experiments were repeated to determine the settling time by allowing samples to settle for different durations (30, 60, 90, and 120 min) using the results of the shaking time tests. Afterward, the results of shaking time and settling time were used as constant conditions for evaluating the influence of silica sand dosage, in which different dosages *i.e.*, 20, 40, 60, 80, and 100 g (to be precise, 166, 333, 500, 666, and 833 g/L) were tested. After each run, the percentages of NH₃-N, iron and zinc removal were measured.

2.5. Absorption Isotherms

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The amount of adsorption at equilibrium state, $q_e \pmod{L^{-1}}$, was calculated by the following equation:

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

where C_o and C_e (mg L⁻¹) are the liquid-phase concentrations of the sample at initial and equilibrium, respectively, W (g) is the mass of composite media used, and V (L) is the volume of the solution. Langmuir and Freundlich, the most common isotherm models, were employed in this study. According to Benefield *et al.* [25], the Langmuir isotherm is based on the assumption that the adsorbed layer will be one thick (homogeneous) molecule, whereas the Freundlich isotherm assumes that the adsorbent has a heterogeneous surface composed of different classes of adsorption sites.

The Langmuir isotherm linear equation is represented by the following equation:

$$\frac{1}{q_{e}} = \frac{1}{K_{a}q_{m}C_{e}} + \frac{1}{q_{m}}$$
(2)

The Freundlich isotherm linear equation is commonly represented by the following equation:

$$\log q_e = \log K_F + \frac{1}{n} \log C_e$$
(3)

The Langmuir constants q_m (mg/g) and K_a (L/mg) are related to maximum adsorption capacity and energy of adsorption, respectively, whereas K_F (mg/g)(L/mg)^{1/n} and 1/n are Freundlich constants related to adsorption capacity and adsorption intensity of the adsorbent, respectively.

3. Result and Discussion

3.1. Pulau Burung Leachate Characteristics

Table 3 illustrates the characteristics of the leachate at PBLS. The table indicates that ammoniacal nitrogen, iron, zinc COD, color, and BOD₅ were present in sizeable quantities with great concentrations of approximately 2,090 mg/L, 7.2 mg/L, 4.7 mg/L, 3,333 mg/L, 3,066 Pt-Co, and 305 mg/L, respectively. Pulau Burung landfill leachate is categorized to be in the phase of methane fermentation, which can be considered as stabilized leachate [5]. In this phase, the high concentrations of color and COD are mainly contributed by dissolved organic substances; however, the presence of high amounts of NH₃-N are attributed to the degradation of nitrogenous compounds present in the dumped solid wastes. In general, ammonia represents the major form of nitrogen in stabilized leachate, constituting over 70% of the Total Kjeldahl Nitrogen (TKN). Zouboulis *et al.* [26] reported that the elevated pH values (7.3–8.8) of stabilized (old) leachate result in an inevitable reduction in the solubility of metals. Also, low metal content could be attributed to adsorption and precipitation processes involving the co-existing sulfides, carbonates, or hydroxide anions. The characteristics of stabilized leachate are extremely different from those of domestic wastewater. Consequently, stabilized leachate is more similar to heavily contaminated industrial wastewaters, indicating that biodegradation processes are not efficient for treatment of such leachate.

Parameter	Minimum	Maximum Average		Standard	Standard	
				Deviation	Discharge Limit ¹	
pH	8.20	8.31	8.26	0.05	6.0–9.0	
Ammoniacal nitrogen, (mg/L)	2010	2090	2050	39.66	5.0	
Iron (mg/L)	3.5	7.2	5.3	1.60	5.0	
Zinc (mg/L)	2.3	4.6	3.4	0.95	2.0	
Copper (mg/L)	0.40	0.70	0.55	0.13	0.2	
Manganese (mg/L)	0.20	0.30	0.23	0.05	0.2	
Nickel (mg/L)	0.20	0.40	0.28	0.09	0.2	
Lead (mg/L)	0.20	0.40	0.32	0.09	0.1	
COD (mg/L)	3027	3333	3180	142.93	400	
Color (Platinum unit, Pt-Co)	2800	3066	2933	116.12	100	
$BOD_5 (mg/L)$	157	305	231	64.56	20	
Suspended solids (mg/L)	70	190	130	50.57	50	

Table 3. Characteristics of the leachate at the Pulau Burung landfill site (PBLS).

¹ Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, under the Laws of Malaysia-Malaysia Environmental Quality Act 1974 [27].

3.2. Optimum Conditions

The optimization of the media performance was achieved by monitoring the influence of one factor at a time on experimental response. This optimization is called the one-variable-at-a-time method. Where only one variable is varied, the others are maintained at a constant level [28]. Figures 1 to 3 demonstrate the experimental results for determining the optimum shaking time, settling time, and silica sand dosage, respectively. The figures also illustrate the influence of each factor on ammoniacal nitrogen, iron, zinc removal efficiency, and pH. As noted in Figure 1, more than 44% of ammoniacal nitrogen, 42% of iron, and 38% of zinc removal was obtained with an optimal shaking time of 90 min, and a settling time of 120 min, shaking speed of 350 rpm, and silica sand dosage of 40 g (333 g/L). As shown in Figure 1, shaking time has a great influence on the parameter removal efficiency; sufficient shaking time is necessary to achieve excellent adsorption of pollutants. Figure 2 shows that the parameter removal increased with increasing settling time up to 60 min, and then remained constant. In fact, the improved removal efficiency at higher agitation speed and with a longer shaking time is due to the rigorous mixing and longer solid-liquid contact bringing about improved interaction between the silica sand and the leachate. The influence of silica sand dosage on the parameter removal efficiencies is shown in Figure 3. Approximately 50.9%, 44.4%, and 39.2% of ammoniacal nitrogen, iron, and zinc, respectively, were removed when 60 g (0.5 kg/L) of silica sand was used (Figure 3). However, with this silica sand dosage, the pH of the treated effluent ranged from 3 to 4, indicating that pH adjustment is strongly required before effluent discharge in order to comply with the Malaysian standard discharge limit. In general, the precipitation of solids increased at lower pH values [29]. Kadirvelu and Namasivayam [30] reported that the removal of iron under acidic conditions was better than under alkaline conditions. Based on the results, the performance of silica sand was shown to be better than activated carbon in terms of NH₃-N removal efficiency from landfill leachate. Whereas approximately 40% of NH₃-N with an initial concentration of more than 1,000 mg/L can be removed by either activated carbon or a mixture of carbon with limestone at a mixture ratio of 5:35 [31], more than 92% removal of NH₃-N can be achieved by employing an ion exchange technique [3]. The ion exchange process appears to be an efficient method among the physicochemical processes for removal of inorganic substances from landfill leachate. However, a considerable disadvantage hindering the success of ion exchange is the high cost of ion exchange resin compared to silica sand.

Figure 1. Influence of shaking time on ammoniacal nitrogen, iron and zinc removal efficiency (dosage = 40 g; shaking speed = 350 rpm; settling time = 120 min).



Figure 2. Influence of settling time on ammoniacal nitrogen, iron and zinc removal efficiency (dosage = 40 g; shaking speed = 350 rpm; shaking time = 90 min).



Figure 3. Influence of silica sand dosage on ammoniacal nitrogen, iron and zinc removal efficiency (shaking speed = 350 rpm; shaking time = 90 min; settling time = 60 min).



3.3. Adsorption Isotherms

Langmuir and Freundlich adsorption isotherm models, which have been profitably applied to many adsorption processes [1,15], were used to study the silica sand adsorption behavior. Figures 4 to 6 show the linear plot of Langmuir and Freundlich isotherms for the adsorption of ammoniacal nitrogen, iron, and zinc, respectively, onto silica sand.

Figure 4. Adsorption isotherm for ammoniacal nitrogen onto silica sand: (a) Langmuir, and (b) Freundlich.





Figure 5. Adsorption isotherm for iron onto silica sand: (a) Langmuir, and (b) Freundlich.

Figure 6. Adsorption isotherm for zinc onto silica sand at: (a) Langmuir and (b) Freundlich.



The empirical constant values of Langmuir and Freundlich isotherms were calculated from the linear plot illustrated in Table 4. As shown in Table 4, the maximum monolayer adsorptions of NH₃-N, iron, and zinc onto silica sand were 6.097, 0.0109, and 0.0046 mg/g, respectively. The coefficient of determination (R^2) was 0.702, 0.728, and 0.612 for ammoniacal nitrogen, iron, and zinc, respectively.

Table 4. Langmuir and Freundlich isotherm constants for adsorption of ammoniacal nitrogen, iron and zinc onto silica sand.

Adsorbate	Langmuir			Freundlich		
	Q(mg/g)	b(L/mg)	\mathbf{R}^2	*K _F	1/n	\mathbf{R}^2
Ammoniacal-nitrogen	6.0975	0.00243	0.702	0.04426	0.719	0.764
Iron	0.0109	0.56574	0.728	0.0107	1.305	0.786
Zinc	0.0046	0.82729	0.612	0.00752	1.309	0.636

*Unit of K_F was (mg/g) (mg/L)ⁿ

The characteristics of Langmuir isotherm can be explained by using a dimensionless equilibrium parameter (R_L), by using the following equation:

$$R_L = \frac{1}{1 + bC_0} \tag{4}$$

where *b* is the Langmuir constant and C_0 is the initial parameter concentration (mg/L). This equation denotes the value of R_L , which indicates whether the isotherm is favorable ($0 < R_L < 1$), unfavorable ($R_L > 1$), linear ($R_L = 1$), or irreversible ($R_L = 0$). By using this equation, the R_L values for ammoniacal nitrogen, iron, and zinc are shown to be in the range of 0 to 1. Consequently, the results indicate that the Langmuir isotherm is favorable for this study. In light of Table 4, the constant R^2 values obtained using the Freundlich isotherm model for ammoniacal nitrogen, iron, and zinc were 0.764, 0.786, and 0.636. The constant 1/n functions as the strength of the adsorbent. The value of 1/n closer to 1 has a high adsorption bond; however, high values of 1/n > 1 show that the adsorption bond is weak [28]. This means that the value of q_e consumes large dosages for a small change in C_e . In this study, the values of 1/n for ammoniacal nitrogen, iron, and zinc were 0.719, 1.305, and 1.309, respectively. Aziz *et al.* [32] reported that when 1/n > 1, the sorption constant grows with the increasing concentration of solution. According to the observed R^2 values obtained from the Langmuir and Freundlich isotherm models, the Freundlich isotherm proved to be a better fit for adsorption because all the R^2 values were higher than those obtained from the Langmuir isotherm.

4. Conclusions

In this study, the adsorption efficiency of ammoniacal nitrogen, iron, and zinc from the semi-aerobic stabilized landfill leachate via natural silica sand as a low-cost adsorbent was investigated. Based on the experimental results, the optimum conditions for batch adsorption were established to be 90 min of contact time, 60 min of settling time, and a shaking speed of 350 rpm. Both Langmuir and Freundlich isotherm studies exhibited that silica sand shows favorable adsorption for the removal of ammonia nitrogen, iron, and zinc in semi-aerobic stabilized landfill leachate. In accordance with the coefficient of determination, ammoniacal nitrogen, iron, and zinc adsorption was better fitted to the Freundlich model. The results indicate that silica sand is potentially efficient as an alternative low-cost adsorbent for stabilized leachate treatment. However, in light of its insufficient effectiveness in terms of removal of all pollutants present in stabilized leachate, particularly organic substances, silica sand can more effectively be used as a filter medium in a preliminarily treatment stage prior to more efficient physicochemical processes.

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