A Batch Study on Removal of Heavy Metals Using Laterite Soil-Pressmud in Landfill Leachate

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ABSTRACT
This batch study focuses on suitability of laterite soil-pressmud as daily soil cover of landfill. The laterite soil samples were mixed with waste from sugar refinery process, namely pressmud at different percentages of weight ratio (10, 30 and 50%). The batch equilibrium tests were carried out and glaringly showed that the laterite soil-pressmud mixtures have the capability to remove more than 62% of Cr, Cu, Mn, Ni, Pb and Zn concentration in leachate. Meanwhile, the removal efficiency of heavy metals from leachate in the laterite soil alone was lower than 50%. Pressmud alone however showed more than 53% removal. The laterite soil-pressmud mixtures, particularly at 30 and 50 percent of pressmud signify great potential as daily soil cover in reduction of heavy metals migration in landfill leachate.

INTRODUCTION
Most landfills in Malaysia use laterite soil alone as a soil cover in their operation. Soil alone may not be an ideal landfill cover material as it cannot reduce major problem such as leachate infiltration. Laterite soils are residual soils, mainly found in tropical regions and generally comprise substantial amount of iron and aluminum oxides [1]. Laterite is a red tropical soil in which the oxides are derived from rock weathering under strongly oxidizing and leaching conditions [2].

The availability of appropriate local soils is an important consideration in any landfill design. Physical and chemical properties of soils are important to a successful engineered landfill design. Disposal facility in landfill is crucial to ensure the environmental safety in term of maintaining the volume of waste and further reduce the potential migration of pollutants into the surrounding geologic media or groundwater. Soils are currently used for the containment of waste, as an interim step while final remediation alternatives are developed, and in coordination with treatment technologies [3]. Soil cover serves as hydraulic barriers [4]. Soil cover with low permeability can reduce the filtration of leachate through waste thus minimizing the migration of toxic pollutants, including heavy metals [5]. It also has been studied that municipal solid waste biostabilization was related to intermediate soil cover characteristics that controlled the decomposition rate of waste [6].

Leachate must be treated prior to discharge and it must meet the discharge limits of treated effluents. Leachate treatment is very complicated, expensive and generally requires multiple processes [7]. Many factors need to be considered when designing the leachate treatment system. Leachate treatment is required during landfill operation and after landfill closure. During the life cycle of the landfill, leachate characteristics will change, so an improvement in treatment system may be required. It has been proved that the use of mixed soil improved leachate quality [8], thus indicating the importance of selecting proper materials as landfill daily soil cover.

This research investigates and evaluates the ability of pressmud (sugar manufacturing waste) mixed with laterite soil to reduce and minimize the migration of heavy metals in landfill leachate. It involves samples

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Materials and Methods

Preparation of Materials

Laterite soil used as daily soil cover in landfills was sampled in Nibong Tebal area and leachate was collected from Pulau Burung Landfill in Penang; while pressmud was sampled from Malaysian Sugar Manufacturing (MSM) Sdn. Bhd., a sugar mill in Seberang Perai, Penang.

The laterite soil and pressmud collected were air-dried and sieved through 200 mm sieve to remove large and coarse pebbles. The laterite soil samples and pressmud were then dried and analyzed for their characteristics. Raw leachate was collected from Pulau Burung Landfill. All leachate samples collected were kept in High Density Polyethylene (HDPE) bottles and preserved at approximately 4°C temperature in a refrigerator. The leachate was then analyzed for its heavy metal concentrations by using Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES, Model Perkin-Elmer Optima 7000).

Basic characterizations of this sample such as pH, specific gravity and moisture content were analyzed. The soil samples were mixed with the pressmud with the percentages of 10, 30 and 50% in weight, respectively. These soil-pressmud mixtures were designated as Pressmud 10 (PM10), Pressmud 30 (PM30) and Pressmud 50 (PM50) accordingly are based on the percentage of the pressmud in the soil. The samples were then ground in the rotary blender in order to obtain a homogeneous mixture. After mixing, all the samples were kept in sealed plastic bags for further analysis.

Batch Equilibrium Test (BET) was performed in order to evaluate the removal efficiency of heavy metals such as Cr, Cu, Mn, Ni, Pb and Zn using soil sample and soil-pressmud mixture (PM10, PM30 and PM50). At the same time, the adsorption capability of the materials tested was also determined.

Several heavy metal species were selected based on raw leachate characterization in which their concentrations were significantly high. These species were usually encountered in leachate from landfills. In this study, the initial concentrations of heavy metals like Cr, Cu, Mn, Ni, Pb and Zn were analyzed by using ICP-OES with their concentrations of 0.62, 0.58, 0.32, 0.34, 0.32 and 2.51 mg/L, respectively.

In this experiment, leachate with various initial concentrations of heavy metals were mixed with the materials (pressmud, mixed laterite soil pressmud and laterite soil alone) at ratio 10:1 (10 mL solution and 1 g of sample) and shaken in a tube for 24 hours in accordance to standard method [9]. The horizontal shaker was used to shake the samples. After reaching equilibrium, the tubes were centrifuged at 5,000 rpm for 25 minutes to separate the liquid and solid form. The supernatant was filtered with Whatman filter paper (No. 42) and then analyzed by ICP-OES.

From these analyses, the concentrations of heavy metals left in the filtrate were used to calculate the amounts of heavy metals absorbed by the pressmud, soil-pressmud mixtures and laterite soil alone. The removal percentage of heavy metals from initial concentration \(C_i\) in leachate was calculated from the following Equation (1). Adsorption capacity and percent removal were used to optimize the material conditions:

\[
\text{Removal} = \frac{C_i - C_e}{C_i} \times 100
\]

Where,

- \(C_i\) = initial concentration of the solution (mg/L)
- \(C_e\) = the equilibrium concentration left in the solution (mg/L)

Results and Discussion

Concentration of Heavy Metals in Pulau Burung Landfill Leachate

Table 1 shows the concentration of heavy metals content in leachate from Pulau Burung Landfill. From the obtained results, Zn showed the highest concentration which was 2.51 mg/L. Concentration of Mn, Ni, Cr, Pb and Cu showed 0.32, 0.34, 0.62, 0.32 and 0.58 mg/L, respectively. The heavy metal concentration for Cu and Zn are slightly higher compared to the data sampling in 2006 [10]. This may be due to the age of the landfill that influenced the heavy metals concentrations. The age of this landfill at the time of the study was almost 34 years since its operation started in 1980. All the heavy metal concentrations exceed the maximum permissible concentration (MPC) limits.

Landfill leachate is a complex wastewater and its composition and concentration of contaminants are influenced by many factors such as the type of waste deposited and the age of landfill [11].
TABLE 1. Heavy metals concentration in Pulau Burung Landfill leachate

<table>
<thead>
<tr>
<th>Heavy Metals</th>
<th>Concentration of Heavy Metals, mg/L</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chromium, Cr</td>
<td>0.55-0.7</td>
</tr>
<tr>
<td>Copper, Cu</td>
<td>0.48-0.86</td>
</tr>
<tr>
<td>Manganese, Mn</td>
<td>0.19-0.66</td>
</tr>
<tr>
<td>Nickel, Ni</td>
<td>0.18-0.54</td>
</tr>
<tr>
<td>Lead, Pb</td>
<td>0.18-0.61</td>
</tr>
<tr>
<td>Zinc, Zn</td>
<td>0.93-3.5</td>
</tr>
</tbody>
</table>

Characteristics of Laterite Soil

According to the British Standard Method (BS 1377-1990), basic properties of the laterite soil samples are shown in Table 2. The pH value shows 4.42 which is acidic and can be considered as strongly acidic condition [12]. The specific gravity of soil is 2.24 while the moisture content showed only 18.4%.

TABLE 2. Basic properties of Laterite Soil

<table>
<thead>
<tr>
<th></th>
<th>Minimum</th>
<th>Maximum</th>
<th>Average</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH value</td>
<td>4.4</td>
<td>4.45</td>
<td>4.42</td>
</tr>
<tr>
<td>Specific gravity</td>
<td>2.21</td>
<td>2.25</td>
<td>2.24</td>
</tr>
<tr>
<td>Moisture Content (%)</td>
<td>18.21</td>
<td>18.51</td>
<td>18.4</td>
</tr>
</tbody>
</table>

Removal Efficiency of Heavy Metals

In this study, initial concentration of several heavy metals in raw leachate such as Cr, Cu, Mn, Ni, Pb and Zn were 0.62, 0.58, 0.32, 0.34, 0.32 and 2.51 mg/L respectively. The removal efficiency of laterite soil, pressmud, PM10, PM30, and PM50 were investigated. Figures 1 to 5 show the results of the removal efficiency of the samples.

Figure 1 depicts the removal percentage of Cr in leachate. From the figure, it can be clearly seen that laterite soil alone could only remove 22.4% of Cr. The addition of pressmud to laterite soil clearly improved the removal efficiency. The removal efficiency of Cr for PM10, PM30 and PM50 were 83.6, 80 and 78.1%, respectively. However, pressmud alone could only remove 72.5% but the value was higher than that of laterite soil alone. From an analysis of variance (ANOVA), it was proved that addition of pressmud as admixture in laterite soil significantly removed Cr in the leachate (p <0.05). Pressmud alone could only remove 69%, much higher than that of laterite soil. Pressmud and laterite soil-pressmud mixture has a tendency to remove more than 72% of Cr in the removal efficiency test. This may be due to the characteristics of pressmud that become sticky when it is drenched and the porosity of the laterite soil-pressmud mixtures is enhanced.

Figure 2 shows the percentage of Cu removal in leachate. From the figure, it can be seen that laterite soil alone could only remove 44% of Cu. The removal efficiency of Cu for PM10, PM30 and PM50 were 75.2, 90.3 and 86.1%, respectively. Pressmud alone could only remove 69%, much higher than that of laterite soil. ANOVA analysis proved that the addition of pressmud as admixture in laterite soil significantly reduced Cu in the filtrate (p <0.05). Apart from the characteristic of pressmud alone which easily turns sticky, the higher of CEC value of pressmud also increased the capability to adsorb heavy metals. It can be said that the addition of pressmud in laterite soil removes higher concentration of Cu from leachate.

Figure 3 depicts the removal percentage of Mn in leachate. From the figure, it can be seen that laterite soil alone could only remove 24.7% of Mn whereas pressmud removed 53.6%. It is clearly shown that the value of pressmud was higher than that of laterite soil alone. From an analysis of variance (ANOVA), it was proved that addition of pressmud as admixture in laterite soil significantly improved Mn concentration (p <0.05). Pressmud and laterite soil-pressmud mixture has a tendency to remove more than 72% of Mn in the removal efficiency test. This may be due to the characteristics of pressmud that become sticky when it is drenched and the porosity of the laterite soil-pressmud mixtures is enhanced.

Figure 4 shows the removal percentage of Ni in leachate. From the figure, it can be seen that laterite soil alone could only remove 24.7% of Ni whereas pressmud removed 53.6%. It is clearly shown that the value of pressmud was higher than that of laterite soil alone. From an analysis of variance (ANOVA), it was proved that addition of pressmud as admixture in laterite soil significantly improved Ni concentration (p <0.05). Pressmud and laterite soil-pressmud mixture has a tendency to remove more than 72% of Ni in the removal efficiency test. This may be due to the characteristics of pressmud that become sticky when it is drenched and the porosity of the laterite soil-pressmud mixtures is enhanced.

Figure 5 shows the removal percentage of Pb in leachate. From the figure, it can be seen that laterite soil alone could only remove 24.7% of Pb whereas pressmud removed 53.6%. It is clearly shown that the value of pressmud was higher than that of laterite soil alone. From an analysis of variance (ANOVA), it was proved that addition of pressmud as admixture in laterite soil significantly improved Pb concentration (p <0.05). Pressmud and laterite soil-pressmud mixture has a tendency to remove more than 72% of Pb in the removal efficiency test. This may be due to the characteristics of pressmud that become sticky when it is drenched and the porosity of the laterite soil-pressmud mixtures is enhanced.
significantly improved the sorption capacity of the soil where no desorption of Mn seemed to occur.

**Figure 3.** Percentage of Mn removal from the solution

Figure 4 shows the removal percentage of Ni in leachate. From the figure, it is clearly shown that laterite soil alone could only remove 46.9% of Ni. Addition of pressmud to laterite soil clearly improved the removal efficiency. The removal efficiency of Ni for PM10, PM30 and PM50 were 78.7, 88.4 and 84.5%, respectively. Pressmud alone removed only 56.9% but it was still higher than that of laterite soil alone.

From ANOVA analysis, it was proved that the addition of pressmud as admixture in laterite soil significantly improved leachate quality (p<0.05). The presence of carboxyl groups in laterite soil-pressmud mixture is believed to be responsible for the sorption of metals ions. There are positive correlations between the removal efficiency and characterization of laterite soils and laterite soil pressmud mixture especially cation exchange capacity (CEC) of that materials. The higher the CEC value of the materials, the higher the percentage removal of the heavy metals observed.

**Figure 4.** Percentage of Ni removal from the solution

Figure 5 shows the percentage of Pb removal in leachate. From the result, PM10, PM30 and PM50 showed higher percentage of Pb removal with 83.8, 93.4 and 92.7%, respectively. All the mixtures indicated more than 83% removal while laterite soil showed the least removal which was at 37.9%. Pressmud alone was capable in removing 93.8% of Pb. From the above figure, it can be said that the mixture of laterite soil and higher pressmud content resulted in higher removal of Pb in leachate. Addition of pressmud as admixture in laterite soil significantly decreased the mobility of Pb from the leachate (p <0.05) in ANOVA analysis.

**Figure 5.** Percentage of Pb removal from the solution

**CONCLUSIONS**

It can be suggested that addition of pressmud to laterite soil significantly reduce the heavy metals concentration in leachate filtrate. As a conclusion, laterite soil-pressmud mixtures have shown a good potential to be used as a daily cover material in landfill as the combinations may help in improving leachate quality and minimizing heavy metals mobility from landfill.

**ACKNOWLEDGMENT**

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**REFERENCES**


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Persian Abstract

چکیده

این مطالعه یک بر روی مناسب بودن خاک لاتریتی به عنوان پوشش روزانه خاک دفن زباله، تمرکز دارد. نمونه های خاک لاتریتی که با ضاکیعات از صنایع پالایشگاه فندک، مه تهیه می شوند نشان داد که حتی ده میلیون تن و نیم تن نیکل، پتاس، روی و شیروی را به عنوان pressmud حذف می کنند. در تجربیات نشان داد که مخلوط pressmud، به خصوص در حالت خاک لاتریتی نسبت به نیکل، پتاس و روی در شیرابی را دارد. در همین حال، روند حذف فلزات سنگین از شیرابی در خاک لاتریتی نسبت به نیکل، پتاس، روی و شیروی در نهایت، pressmud به آنها درصد می دهد. به طور کلی نشان می دهد که خاک لاتریتی به عنوان پوشش روزانه خاک دفن زباله محل دفن زباله را دارد.