The potential of semi-permeable bentonite and zeolite composite on the reduction of Pb (II) concentration in landfill

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Abstract. An alternative material that could be used as a liner, which can avert the heavy metal migration from landfill to soil, is the combination between bentonite and zeolite. This study aims to analyze the Pb2+ adsorption capacity of the liner and permeability of bentonite with the addition of zeolite in several percentages. There were four different types of composite in the experiment, such as pure bentonite (BZ0), bentonite + zeolite 2% (BZ2), bentonite + zeolite 5% (BZ5), and bentonite + zeolite 8% (BZ8). Bentonite and zeolite composite was inserted into a cylindrical reactor, which was compacted to adjust the condition of the study to be similar with existing condition in the landfill. This study was conducted until the leachate produced and lead concentration was tested using samples from the liner and leachate in the outlet. The results showed that the highest efficiency of lead reduction was obtained from BZ8 with the percentage of 95.2%, while the highest permeability coefficient was found in BZ8 with the value of 7.76 x 10^{-11} cm/s. The addition of zeolite has been proven to be able to reduce the lead concentration of leachate, but it increased the possibility of leakage.

1 Introduction

In urban waste management, landfilling is a method that is commonly used as a place for the final processing of waste. However, there is a very important environmental problem to consider, namely the presence of leachate. Leachate is formed due to the process of decomposition of waste, which is supported by the presence percolation and infiltration of rainwater through the waste [1]. Landfill leachate is a complex pollutant that has a high concentration of dissolved organic matter [2]. In the leachate, there are various materials that are harmful to the environment, such as phenols, aromatic compounds, ammonium, and heavy metals [3].

Heavy metal contamination illustrates that serious environmental pollution is happening. One of heavy metal that its negative impact on the environment has been well known is lead, which has a high toxicity level and is difficult to process [4], [5]. In addition, lead also causes impacts on humans, such as internal organ damage, behavioral disorders, mental disorders, cancer, and nerve damage [6]–[8]. Therefore, it is necessary to have a semi-permeable layer that is able to hold the leachate so that it cannot escape and prevent environmental pollution [9].

There are various materials that can be used as landfill liners which have a function as barriers to hold leachate out of landfill cells into the surrounding environment. One of commonly used material as a landfill liner is compacted clay, because of its abundant availability and low price [10]–[12]. Compacted clay is able to remove lead (Pb2+) in an adsorption process due to the presence of negative ions contained in it [13]. Bentonite, which is included in compacted clay, is often chosen because it has high porosity, high cation exchange capacity, large surface area, and small particle size so that it can adsorb more pollutants [14]–[17]. Bentonite can be found in both natural and sedimentary soils [18]. Bentonite mainly consists of montmorillonite, with an arrangement of tetrahedral sheets from SiO4 which coincide with an octahedral sheet of Al3+ (Fe2+/Mg2+) [10], [19]. However, there are some problems with the use of pure bentonite as a liner, such as shrinkage and low shear stability [20], [21].

Zeolite is a material that can be used to improve the shear stability of a liner due to the nature of zeolite as a soil stabilizer. In its use, zeolite is widely used in construction activities. The presence of crystalline in zeolite is able to adsorb various particles that have smaller size than its diameter [21]. Zeolite can also increase the adsorption capacity of the liner because zeolite has a cation exchange capacity of 200 to 400 meq/100 grams, which is almost the same as the capacity possessed by bentonite [22]–[24]. Removal of various heavy metal compounds such as lead, cadmium, zinc, and manganese can be accommodated by using adsorbents from zeolite. However, zeolite has disadvantages because it can increase the permeability of
the liner mixture, so the composition of the mixture of bentonite-zeolite must be considered [25].

To date, research on the use of bentonite and zeolite composite materials has been carried out in wastewater remediation, while its use in eliminating lead as well as landfill liners has not been much studied. In this study, the performance of lead reduction (Pb\(^{2+}\)) on bentonite-zeolite composite landfill liners was investigated. In addition, the percentage of removal from lead and increase in permeability values are discussed.

2 Methodology

Leachate from Jatibarang Landfill was used. It was taken from the Leachate Treatment Plant. Pb\(^{2+}\) test conducted after in Diponegoro University. The initial concentration from the leachate was 1.584 mg/L. This study used standard from Peraturan Daerah Jawa Tengah No. 5 Tahun 2012, which has the maximum lead concentration of 0.1 mg/L hence it will be the target of final lead concentration. This study was conducted until there are leachate that came out from the effluent of the reactor, which happened in about 30 days.

Bentonite material used in this study was obtained from mining minerals and bentonite rocks in Giriwoyo District, Wonogiri Regency, Central Java, with a mesh size of 0.075 mm. Bentonite is a pale-colored plastic material that can expand when there are water addition. It turns into pulp if there is too much water while if it is dry, a crack will be formed [26]. Meanwhile, zeolite was obtained from mineral and coal mining in Wangon District, Cilacap Regency, Central Java, in the form of granules with 2 mm mesh.

Four (4) acrylic-based reactors were built, with a PVC base. The reactor has a total height of 11 cm (8 cm acrylic and 3 cm PVC) and a diameter of 10 cm. Each reactor was given a liner from bentonite and zeolite mixture with different variations, which were pure bentonite (B0), bentonite with the addition of zeolite 2% (BZ2), bentonite with the addition of 5% zeolite (BZ5), and bentonite with the addition of zeolite 8% (BZ8). Leachate samples that have been obtained were inserted into each reactor as much as 30 ml. Pressure was applied to each reactor by 4 kPa, which is the pressure of waste in the Jatibarang Landfill and provide similar conditions as the existing. After the leachate flowed through the landfill liner, landfill liner then divided into three layers (top, middle, and bottom layers).

The Pb\(^{2+}\) concentration test was carried out onto the leachate which coming out of the reactor outlet and each layer of the liner. The effluent leachate samples were taken by spilling the leachate in the reactor into a container to separate it from the liner. Next, the liner is taken using a spoon with the same depth of 1 cm in each layer. After leachate sampling, Pb\(^{2+}\) concentration test carried out using SNI 06-6989.8-2004 method with Atomic Absorption Spectrophotometry (AAS) [27]. Meanwhile, to determine the characteristics of the landfill liner material, optimum water content (OMC) and maximum dry density (MDD) test was carried out by conducting a Standard Proctor Test based on ASTM D-698 [28]. Permeability Coefficient of each bentonite-zeolite composite from each reactor was carried out using the Falling Head method based on ASTM D5084-03 [29]. All tests were carried out in duplicate at the Department of Environmental Engineering, Faculty of Engineering, Diponegoro University.

3 Results and Discussion

3.1 Lead Concentration

Measurement of lead concentration in effluent landfill leachate was conducted to examine the lead content in the leachate that came out from the reactor and in the bentonite and zeolite composite liners. From these results, it can be estimated the efficiency of lead removal and lead content entering the environment and compared to existing quality standards, which is no more than 0.01 mg/L.

1.1.1 Lead Concentration in Leachate Effluent

Lead concentration in the leachate coming out into the environment needs to be determined to prevent a decrease in the quality of the surrounding environment. Table 1 shows the concentration of lead in the leachate coming out of the reactor.

| Sample | Lead Concentration (mg/L) | Removal Efficiency (%) |
|--------|---------------------------|------------------------|
| B0     | 0.503                     | 68.2                   |
| BZ2    | 0.397                     | 74.9                   |
| BZ5    | 0.189                     | 88.1                   |
| BZ8    | 0.076                     | 95.2                   |

Based on table 1, the concentrations at B0, BZ2, BZ5, and BZ5 were 0.503 mg/L; 0.397 mg/L; 0.189 mg/L and 0.076 mg/L respectively. This indicates that with the addition of zeolite, there is a greater decrease in lead concentration. Meanwhile, only bentonite and zeolite composites with the addition of 8% zeolite met the quality standard, which was less than 0.1 mg/L. On the other hand, the efficiency of lead removal in each reactor can be calculated. Lead removal was greater following the increasing number of zeolites in the liner, where B0 has a removal efficiency of 68.2%; BZ2 of 74.9%; BZ5 of 88.1%; and BZ8 of 95.2%. These results are in accordance with Turan [21], which has higher adsorption capacity in the composite with the highest zeolite addition.

Pawar [10] found that the removal percentage of the metal will continuously decrease during the adsorption process, which is because the presence of the sorbing ions is smaller than the availability of active sites. The
decrease will happen until the liner is saturated. Different materials of clay have various sorption capacities. Commercial bentonite has sorption capacity of 50 mg/g [30], while zeolite has higher capacity, which is 75 mg/g [23].

1.1.2 Lead Concentration in each layer of landfill liner

Lead concentration in the landfill liners that have been passed by leachate needs to be determined to investigate the capacity of each landfill liner to remove lead from leachate, which can be seen from the lead content of each landfill liner. The results of testing the lead concentration in a landfill liner that has been divided into three parts are illustrated in table 2.

Table 2. Lead Concentration in Landfill Liner

| Layer    | Concentration (mg/L) |
|----------|----------------------|
|          | B0  | BZ2  | BZ5  | BZ8  |
| Top      | 0.171| 0.464| 0.716| 0.764|
| Middle   | 0.078| 0.369| 0.553| 0.682|
| Bottom   | 0.052| 0.315| 0.480| 0.651|

Table 2 represents B0 having the greatest lead concentration of 0.171 mg/L in the top layer of the landfill liner. As it goes downward, the lead concentration in the liner gets smaller with a lead concentration of 0.078 mg/L in the middle layer and 0.052 mg/L in the lower layer. BZ2, BZ5, and BZ8 also have smaller lead content in the lower layer. BZ2 has lead concentrations in the upper, middle and lower layers of 0.464 mg/L; 0.369 mg/L; and 0.315 mg/L respectively. Lead concentration in the BZ5 landfill liner is 0.716 mg/L in the upper layer, decreasing in the middle and bottom, where the lead concentration are 0.553 mg/L for the earlier and 0.480 mg/L for the latter. Meanwhile, the concentration of lead in the upper layer of BZ8 is 0.764 mg/L, 0.682 mg/L in the middle, and 0.651 mg/L in the lower layer. These results indicate that the increment of zeolite addition caused greater lead concentration in the landfill liner. It is because there is more lead that can be absorbed by landfill liners with greater zeolite content so that the lead concentration in the effluent is smaller in the largest addition of zeolite.

3.2 Coefficient of Permeability

Before determining the permeability of each bentonite-zeolite mixture, an optimum water content (OMC) and maximum dry density (MDD) test was performed using the Standard Proctor Test. To examine these parameters, the mixture was added with 5 different water addition. The obtained results of MDD were varied from 1.1 gr/cm³ to 1.3 gr/cm³ while the OMC were ranged from 35% to 46%, as shown in table 3. Based on these results, it can be determined that the OMC and MDD the best characteristic for OMC was found in B0 (46%) and BZ8 has the best value for MDD.

Table 3. OMC and MDD of Bentonite and Zeolite Composite

| Water Addition (cm³) | OMC (%) | MDD (gr/cm³) |
|----------------------|---------|--------------|
| B0                   | 46      | 1.102        |
| BZ2                  | 42      | 1.146        |
| BZ5                  | 39      | 1.214        |
| BZ8                  | 35      | 1.268        |

Soil permeability is a parameter that indicates the amount of fluid that can flow through porous material. The factor that determines permeability is the size of the soil grain, where the smaller the size of the soil grain, then the possibility of water to pass through the medium is also smaller. The value of permeability of a landfill liner should not exceed 1 × 10⁻⁹ cm/s so that water cannot pass through the layer [31]. The permeability of the liners in each reactor is illustrated in table 4.

Table 4. Hydraulic Conductivity of Landfill Liner

| Sample | Hydraulic Conductivity (cm/s) |
|--------|-------------------------------|
| B0     | 1.12 x 10⁻¹¹                  |
| BZ2    | 1.60 x 10⁻¹¹                  |
| BZ5    | 2.41 x 10⁻¹¹                  |
| BZ8    | 7.76 x 10⁻¹¹                  |

Pure bentonite (B0) is the liner with the lowest permeability with the value of 1.12 x 10⁻¹¹ cm/s. The addition of zeolite by 2% (BZ2), 5% (BZ5), and 8% (BZ8) caused an increase in the permeability coefficient, which become 1.60 x 10⁻¹¹; 2.41 x 10⁻¹¹; and 7.76 x 10⁻¹¹ respectively. Based on these results, the addition of zeolite to bentonite can increase the permeability of the mixture of the two materials. However, this increase in hydraulic conductivity is still acceptable to be a basic material for landfill liners because it is still below the specified maximum value.

4 Conclusion

Bentonite and zeolite are landfill liner materials that can reduce the concentration of lead (Pb²⁺) found in leachate. Composite mixtures of the two materials used in this study, which were pure bentonite (B0), bentonite added with 2% zeolite (BZ2), bentonite added with 5% zeolite (BZ5), and bentonite added with 8% zeolite (BZ8). These mixtures were put into acrylic reactors, which have a diameter of 10 cm, as a landfill liner with a height of 10 cm, as a landfill liner with a height...
of 3 cm. The results of the study show that B0 has the lowest lead removal with 68.2% efficiency while BZ8, which contains zeolite higher than the other, has the highest efficiency with the value of 95.2%. Hence, the mixture of the two materials can increase the adsorption capacity of the landfill liner. On the other hand, BZ8 has the highest hydraulic conductivity among all landfill liners with the value of 7.85 x 10⁻¹¹ cm/s but this value still meets the maximum permeability coefficient allowed, which is 1 x 10⁻⁹ cm/s. It can be interfered that the use of higher zeolite composition in the liner is still possible. Research related to the use of various mixing of compacted clay as landfill liner material and provide lead adsorption still have the potential to be explored further.

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