The Effect of Fly Ash Ratio Addition as Layering Material Using Free Draining Column Leach Test Method on Laboratory Scale

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ABSTRACT

Acid mine drainage (AMD) is formed due to the oxidation of sulfide minerals in the presence of water and air. AMD already entering the environment can damage the aquatic ecosystem due to low pH and high dissolved metal concentrations. Efforts to prevent the formation of AMD are by regulating the stockpiling of materials containing sulfide or potential acid forming (PAF) with non-acid forming (NAF) to avoid oxidation reactions of sulfide minerals. The purpose of this study was to simulate the effect of adding fly ash ratio as a PAF rock coating material to the quality characteristics of leachate using the laboratory-scale free draining column leach test (FDCLT) method. Adding fly ash ratio as a layering material significantly affects the quality of the resulting leachate. This is due to the physical condition of the fly ash material, which cannot prevent the meeting of AMD-forming components.

Keywords: Acid Mine Drainage, Fly Ash, Free Draining Column Leach Test.

INTRODUCTION

Coal mining is one of the sectors that provide energy to the world. One is to generate electricity that will be used on an industrial scale. Along with the increase in coal mining operations, it has a negative impact on the environment in the form of the emergence of acid mine drainage (AMD), which can cause long-term damage to waterways and biodiversity because the waste produced contains toxic substances [1].

Acid mine drainage (AMD) results from the oxidation of sulfide minerals by oxygen, producing ions easily soluble in water. Besides being a solvent medium, water is a transport medium for AMD to the environment. The AMD that enters the environment will damage the aquatic ecosystem due to the low pH and high concentration of hazardous metals [2].

There are several methods of AMD prevention. Prevention generally aims to minimize AMD components like oxygen supply, leachate infiltration and discharge, sulfide mineral presence, and bacteria [1]. One of the efforts made is using dry cover [3]. A dry cover is considered the most effective method for minimizing air and water penetration [1].

The prevention of AMD, which is frequently practiced in Indonesia, uses a layering method by arranging the stockpiling of potential acid forming (PAF) material with non-acid forming (NAF) material. The PAF
material is placed below the NAF material, with the expectation that the NAF material will block oxygen supply and water infiltration. This method has proven efficient in the long term in preventing AMD formation [1],[4].

However, the availability of NAF material in terms of quality and quantity in the field tends to be limited, and the amount is much less than PAF material in several coal mines in Indonesia. One effort is to substitute NAF material using fly ash derived from combustion residues in power plants. Fly ash has alkaline chemical characteristics and can be used as a neutralizer [5],[6]. Physical characteristics can be used as a layering material to prevent AMD formation reactions [7], increase pH, and reduce dissolved metal concentrations [8].

Fly ash has suitable characteristics as a layering material because it has a small grain size and alkalinity that can help improve leachate water quality [8]. Fly ash can be used as a PAF rock layering material to increase pH and reduce dissolved metal levels [8]. However, fly ash's physical and chemical conditions are strongly influenced by coal quality, combustion conditions, and the length of time fly ash is exposed to the air when stockpiled.

This study was conducted to determine the effect of the fly ash addition ratio used as a layering material by using the free-draining column leach test (FDCLT) on a laboratory scale. The fly ash material used in this study was taken from a coal-fired power plant ash pile. The fly ash material was placed on top of the PAF rock layer with a ratio of 2:1, 1:1, and 2:3 in the kinetic test column (FDCLT) to determine its effect on the quality of the leach water produced.

**METHODOLOGY**

**Material**

Fly ash samples were taken from the ash pile at one of the power plants in South Kalimantan. The samples were then dried to remove the moisture content in the fly ash. The drying was done by oven at 105–110ºC for approximately 8 hours. The samples were then sieved with a mesh size of -3; this size was used in the study.

**Physical Analysis**

Physical Analyses were conducted to determine the rock's grain size distribution and property index. In addition, scanning electron microscopy energy dispersive x-ray spectrometer (SEM-EDS) tests were conducted to determine the morphology and grain shape of the samples. Analyses were carried out on samples with a size of approximately 100 mesh that had been coated by carbon coating or sputtering to increase the conductivity of the sample for better observation.

**Mineralogy and Elemental Analysis**

Mineralogical and elemental analyses were conducted to determine the mineral and elemental composition of the materials used in this study. The mineralogy analysis used Rigaku SmartLab x-ray diffraction (XRD) with the operating parameters of an X-ray source from a Cu-Kα1 cathode tube with energy of 8.04 keV, the scanning angle 20 used in the range between 5º –65º with an increment of 0.01°. This analysis resulted in diffractometer patterns that were analyzed using SmartLab Studio II software. Elemental analysis was carried out qualitatively and quantitatively using quantitative analysis result-reference intensity ratio. Elemental analysis uses Rigaku Supermini 200
wavelength dispersive x-ray fluorescence (WD-XRF) on -200 mesh and pressed powdered samples, with the form of oxide composition results.

**Static Test**

The static test is carried out to determine the samples’ acid-forming potential, including acid-base accounting (ABA), net acid generation, and paste pH. ABA includes determining the maximum potential acidity (MPA), acid neutralizing capacity (ANC), and net acid production potential (NAPP), which is obtained from the reduction of MPA and ANC. The static test results will be used for the geochemical classification of samples [9].

**Kinetic Test**

The kinetic test is intended to simulate the oxidation and leaching processes of the rock samples. The leached water will be tested to determine the potential for AMD formation. The kinetic test uses the free-draining column leach test (FDCLT) method. The column has a diameter of 15 cm with a height of 30 cm. Before the sample is inserted, filter paper is first placed in the column. Fly ash material and PAF rock will fill the column approximately 25 cm thick. Then, gravel is added on top of the fly ash, with an expectation that the materials will be able to reduce the evaporation process during the dry cycle. A simplified illustration of the column is shown in Figure 1.

This test applies a wet cycle, and watering is performed using distilled water with a soil-water ratio of 2:1. Meanwhile, the dry cycle uses a 60-watt incandescent lamp with a 30º–35º C temperature. This research was conducted using daily, three days, and weekly cycles. Leachate water results were measured using multi-parameters, including pH, total dissolved solids (TDS), electrical conductivity (EC), and oxidation-reduction potential (ORP).

![Figure 1. Illustration of kinetic analysis column.](image)

**RESULTS AND DISCUSSION**

**Physical Condition**

Fly ash material has a perfectly rounded grain shape, and some grains bind to each other (Figure 2). This condition occurs because fly ash material has pozzolanic and cement-like properties. The grains clump together when it is in piles, open places, and exposed to water. Clumped grains in fly ash cause the grain size to be more distributed in silt size by 50.5% and gravel by 17% (Figure 3). Fly ash has a porosity of 57%, which is due to clumped grains.
Minerals and Elements

The mineralogical analysis on fly ash shows mineral composition including quartz (SiO₂), magnetite (Fe₃O₄), sodalite (Na₈(Al₆Si₆O₂₄)Cl₂), hematite (Fe₂O₃), lime (CaO), and periclase (MgO). Based on the results of XRF analysis, fly ash is generally composed of silica (SiO₂), iron (Fe₂O₃), and aluminum (Al₂O₃). Magnesium (MgO) and calcium (CaO), which are alkaline, are found in it.

Meanwhile, the minerals in the PAF sample are composed of quartz (SiO₂), magnetite (Fe₃O₄), kaolinite (Al₂Si₂O₅(OH)₄), sodalite (Na₈(Al₆Si₆O₂₄)Cl₂), and pyrite (FeS₂). The results of XRF analysis on PAF samples show that silica (SiO₂) and aluminum (Al₂O₃) are the dominant elements. Sulfur in the form of sulfur trioxide (SO₃), which is acidic, was present in the PAF sample.

Static Test

The fly ash sample showed high ANC values derived from lime and periclase, while the PAF sample produced acid derived from pyrite (Table 1). Based on the static test, the PAF sample has no neutralizing ability, as indicated by the low ANC value. Considering each sample's NAPP value, an appropriate kinetic test scenario can be determined (Table 2) using a negative NAPP value (no potential to produce acid), with the scenario shown in Figure 3.
Table 1. Static test results.

<table>
<thead>
<tr>
<th>Sample</th>
<th>pH</th>
<th>MPA</th>
<th>ANC</th>
<th>NAPP</th>
<th>NAG</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>FA</td>
<td>0.22</td>
<td>6.74</td>
<td>337.88</td>
<td>-326</td>
<td>9.01</td>
<td>NAF</td>
</tr>
<tr>
<td>PAF</td>
<td>4.02</td>
<td>74</td>
<td>0.0</td>
<td>74</td>
<td>2.60</td>
<td>PAF</td>
</tr>
</tbody>
</table>

Table 2. The scenario of kinetic test.

<table>
<thead>
<tr>
<th>Kode</th>
<th>Kolom</th>
<th>PAF (gr)</th>
<th>Fly ash (gr)</th>
<th>NAPP (kg H$_2$SO$_4$/ton)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>1500</td>
<td>-</td>
<td>74</td>
<td></td>
</tr>
<tr>
<td>S2</td>
<td>-</td>
<td>1500</td>
<td>-326</td>
<td></td>
</tr>
<tr>
<td>S3</td>
<td>1500</td>
<td>750</td>
<td>-59</td>
<td></td>
</tr>
<tr>
<td>S4</td>
<td>1500</td>
<td>1500</td>
<td>-126</td>
<td></td>
</tr>
<tr>
<td>S5</td>
<td>1500</td>
<td>2250</td>
<td>-226</td>
<td></td>
</tr>
</tbody>
</table>

Figure 3. Kinetic test scenario illustration.

**Kinetic Test**

The leachate, collected from the kinetic test, was measured to obtain physical parameter values related to the quality of potential AMD formation. Figure 4 shows the pH of the PAF (S01) and fly ash (S03) control column leachate according to the kinetic test results. The fly ash sample classified as NAF produced an alkaline pH, and the PAF sample was classified as acidic.

The pH of the leachate water at the beginning of the cycle continued to change, and this occurred due to leaching from the initial load flushing (Figure 5). The pH of the leachate water dropped in the 3-day cycle due to the oxidation of calcium that occurred in that cycle. The pH was then observed to be stable until the end of the study.

ORP has an inverse relationship with the pH value. The greater the pH, the smaller the ORP value and even negative. In Figure 6, the ORP value is > 200 mV because the sample undergoes a lot of oxidation, the solvent will lose electrons, and the leach water becomes rich in H$^+$, causing the leach water's pH to become smaller (more acidic).
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TDS describes the amount of dissolved solids in leachate water, which correlates with the amount of dissolved metals. The lower the pH value, the higher the TDS value (Figure 7). At low pH, H⁺ ions will dissolve the metals in the leachate water to increase the pH and the EC. When the TDS value is high, the EC value will be higher because of the dissolved metals that conduct electricity.

Based on the kinetic test results, the fly ash addition ratio does not significantly affect the pH value or other parameters. This is due to the fly ash material that cannot prevent oxidation of the PAF layer. This is evidenced by the pH value of the layering column's leach water, which is still close to the PAF control column. This close pH value is interpreted as pyrite oxidation still occurring.

The physical condition of the fly ash causes the oxidation process in the layering column. Fly ash has grains that clump together, creating gaps in the fly ash layering (Figure 8). During the dry cycle, the water in the fly ash layer will quickly disappear so that oxygen can still enter to oxidize the PAF layer. The neutralizing condition becomes less effective because the fly ash is assumed to have been stockpiled too long, thus reducing its performance as a neutralizer.
CONCLUSION

Ideally, the increase in fly ash ratio will improve the leachate water quality, but this result did not occur in this study due to the physical condition of the fly ash that had clumped together. Clumping makes gaps between fly ash grains, so oxygen can still enter to oxidize the PAF layer during the dry cycle in the kinetic test. The utilization of fly ash as a layering material must consider the physical condition of fly ash. The physical condition of fly ash is essential in preventing the meeting of acid mine drainage forming components. The recommendation is to use fly ash with other materials added on top of the fly ash layer (e.g., topsoil) to prevent oxygen from entering the PAF layer.

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Figure 8. Gaps formed due to fly ash grains clumping together.
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