

## **FUNDAMENTAL RESEARCH OF FLY ASH AND ORGANIC MATERIAL UTILIZATION AS DRY COVER LAYER FOR ACID MINE DRAINAGE PREVENTION METHOD: COLUMN LEACHING STUDY**

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### **ABSTRACT**

Over the past few years, Indonesia has been accounted as one of the biggest countries in producing the coal for energy source. In 2011, Indonesia became the top exporter of coal steam in the world and keep becoming the top five up until now. This has also been accompanied by the promoted domestic coal demand since the government regulations about domestic market obligation and the national energy mix plan in 2030 were issued to increase domestic coal uses. As the result, coal mines in Indonesia are abundant and still increase. Despite the benefit of mining, the growth of mines implies to the growing negative environmental effects caused by mining activity. One of the negative effects is acid mine drainage (AMD). AMD is a severe water pollution issue in the coal mine. AMD occurs because of the sulfide mineral oxidation by oxygen in the presence of water which produces acidity. Consequently, acidic water with elevated concentration of dissolved heavy metals is resulted. As a major issue in the mining, AMD has to be minimized. This is important because AMD heavily affect living organisms and harm the ecosystem by flowing to the surface and groundwater. Moreover, AMD treatment is an expensive as well as complicated matter to be conducted which need a long term management system. Therefore, an AMD prevention method is favorable and needs to be applied on the mine.

The common practice in AMD prevention is a dry cover technique. In this technique, rock that is potential in producing acidity (PAF) will be placed below non-acid producing rock (NAF). However, several issues regarding the material availability could be faced because dry cover is a site specific method that depends on the mine site situation. This research mainly aims to overcome the problems that commonly happen to Indonesia mining site and the world: shortage of non-acid forming rock and the lack capacity of neutralizing capacity. The utilization of additional material cover layer is proposed, by using fly ash and organic material combination. Fly ash is known for its alkaline properties while organic material could consume the oxygen during the degradation process. To investigate the possibility of using these materials, a column leaching test in the laboratory scale was conducted with several scenarios of simulation. The leachate water behavior is observed in the experiment, including the measurement of water quality (pH and EC), major cations-anions, dissolved metal concentration and total organic carbon. The result suggests the application of fly ash and organic material

as cover layer material, especially in the case of mine with domination of PAF rock material. By comparing between column with and without the additional cover layer, the leachate water was improved sufficiently.

**KEYWORDS:** Fundamental Research

## **INTRODUCTION**

The potential of AMD generation is quite high in Indonesia coal mining site. A common problem that exists is the shortage amount of non-acid forming (NAF) rock availability while the potential acid forming (PAF) rock is abundant. It could be worsened when the NAF lacks in the capacity of acid neutralization. The neutralizing capacity of NAF has a significant role in the AMD generation process, by increasing water pH thus precipitating dissolved heavy metals. Both conditions, lack of NAF and insufficient neutralizing capacity, may affect the prevention method strategy since the dry cover conventional method in overburden dumping is ineffective to reduce the AMD generation. Therefore, it is necessary to apply appropriate techniques for overcoming the problem in the dry cover method.

One of the methods to overcome those problems is by utilizing another layer as cover layer that has the capability in improving and/or reducing AMD generation. In this research, study of fly ash and organic material utilization as a cover layer in the dumping area of Indonesia coal mine site will be carried out. Fly ash with the capacity in producing a highly alkaline solution recently has been studied broadly in the world. This material is available mostly in the coal mine, as a residue of coal-steam power plant which mostly located near the mine. However, Indonesia has a restriction of fly ash utilization because of its classification as hazardous waste material. Moreover, the excessive mineral concentration within the fly ash also caused awareness of metals dissolution to the groundwater. Therefore, in order to reduce the amount of fly ash, combination of fly ash and organic material will be studied in this research. Organic material with the oxygen consumption ability during its degradation has a great capability of minimizing AMD generation, with restraining the oxidation of sulfide mineral, as the main process of AMD. Dissolved organic material also could react with sulfate to precipitate metal in the form of metal sulfide with the presence of sulfate-reducing bacteria (SRB).

In order to understand the impact of fly ash and organic material as a cover layer, column leach test with layering scenario was conducted in the laboratory scale. As the study case, Asam-asam mine site that belongs to PT Arutmin Indonesia (PTAI) was chosen for field sampling. This mine site was proven suitable in this case, because of the greater amount of PAF rocks compared to NAF rock which also has lack capacity in acid consumption. During the research, pH, electrical conductivity (EC) and oxidation-reduction potential were directly measured to obtain information of leachate water behavior. Major cation-anion, dissolved metal concentration and dissolved organic carbon were also measured to provide information about leachate behavior.

## **METHOD**

### **Material**

Rock samples are coal-bearing sedimentary rock that obtained on the Asam-asam site. Fly ash was collected from

Asam-asam coal steam power plant that belong to the National Electric Company, while the organic material was obtained from the empty fruit bunches (EFB) of palm fruit oil plantation, the company that is adjacent to the mine site. Rock samples that were categorized as NAF material then uniformly mixed together with the exact same weight and identical grain size. This procedure was carried out due to the consideration that the dumping area has heterogeneous bulk rock material in the site. The same procedure was also conducted to the PAF material. In order to have a better evaluation of cover material effect in improving the leachate water quality, acid producing capacity was increased. It was done by adding pyrite mineral to the PAF mix material in the 2% w/w since pyrite is the major sulfide mineral that caused AMD in the coal mine. The static test was carried out in the laboratory to know the geochemical characterization after mixing the material (Sobek, 1978; AMIRA, 2002). The result of material after mixing, together with fly ash material can be seen in the Table 1.

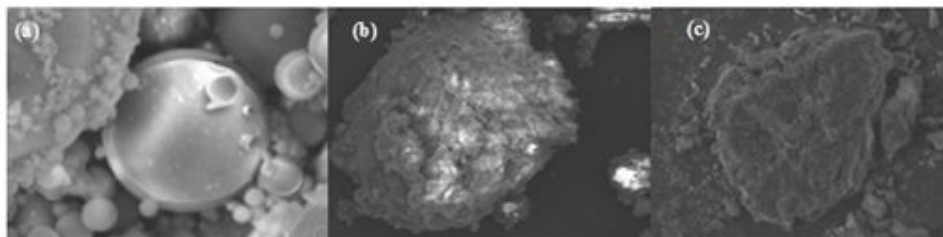
**Table 1: Static Test Results of Material for Column Test**

Sample	pH Paste	TS <sup>a</sup> (%)	MPA <sup>b</sup> ANC <sup>c</sup> NAPP <sup>d</sup>			NAG pH <sup>e</sup>	NAG	
			(kg H <sub>2</sub> SO <sub>4</sub> /ton)				(kg H <sub>2</sub> SO <sub>4</sub> /ton)	
PAF mix	4.16	1.1	33.78	1.95	31.83	2.33	42.65	0
NAF mix	5.77	0.11	3.32	2.65	0.67	6.04	0	5.87
Fly ash	11.81	0.59	17.97	224.69	-	10	0	0

<sup>a</sup> TS – Total Sulfur <sup>b</sup> MPA – Maximum Potential of Acidity <sup>c</sup> ANC – Acid Neutralizing Capacity <sup>d</sup> NAPP – Net acid producing potential <sup>e</sup> NAG – Net acid generating

SEM data of material surface were provided in the Figure 1. Fly ash particle dominantly has a spherical shape with various particle sizes. Meanwhile, PAF and NAF Mix material have heterogeneous shapes and sizes of particle. From the SEM-EDX measurement, major elements on the surface of PAF Mix are O (56.6%), Al (14.1%), Si (27.6%), K (0.46%), Ti (0.49%) and Fe (0.7%) while NAF Mix major elements are O (57.4%), Si (27.47%) and Al (14.2%). Major elements on the surface of fly ash are C (7.1%), O (51.27%), Mg (2.7%), Al (1.4%), Si (9.8%), Ca (3.5%) and Fe (24.08%).

Figure 2 provides the particle size distribution of PAF Mix, NAF Mix and fly ash before the simulation was started. The grain size of PAF and NAF Mix is almost similar which passed the #20 mm sieve (size opening=19 mm) with the d<sub>50</sub> equals to 0.80 mm and 0.75 mm, respectively, while d<sub>20</sub> of both materials equal to 0.20 mm. Moreover, the fly ash particle size is finer than the PAF and NAF Mix particle. It has d<sub>50</sub>=0.21 mm and d<sub>20</sub>=0.08 mm.



**Figure 1: SEM Image of Fly Ash 1000x (a), PAF Mix 2000x (b) and NAF Mix 2000x (c)**

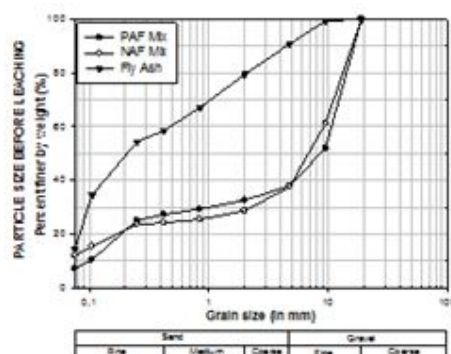


Figure 2: Particle Size Distribution of Material before the Leaching Test

### Method

The leaching simulation was followed a method of Free Draining Column Leach test from AMIRA, with modification. The column that was used in this simulation has diameter 100 mm and total height 300 cm, while the control column per material had diameter 100 mm and total height 100 mm as shown in Figure 3. Layering scenario was selected due to the consideration of mining method that is used in the mine site. Moreover, layering scenario has been proven as an effective method for AMD prevention based on the previous study (Kusuma, 2010).

The simulation consists of column with and without fly ash and organic material cover layer. This was carried out to investigate the effect of cover for the AMD generation. The column with cover had two scenarios of PAF and NAF layer ratio. The purpose is to compare the effectiveness of cover in the case of a different PAF amount. In the control column PAF Mix, NAF Mix and cover layer behavior were investigated separately. The weight of each material of layer is shown in the Table 2.

Leaching test was conducted in total 56 days, with phases of wetting and drying in order to resemble the mining field condition. The drying phase was a daily cycle, consists of 12 hours heating by the lamp (measured temperature in the column,  $T=35\text{--}40^{\circ}\text{C}$ ) and 12 hours natural drying. Spraying by 250 ml of deionized water was conducted for wetting phase and the leachate water collected in the 300ml flask. After the leachate water came out, it directly measured for pH and EC. Then, leachate water was filtered with  $0.45\ \mu\text{m}$  filter and acidified. Major cation-anion measurement was conducted by using Dionex ICS-90 ion chromatography. On the other hand, dissolved metal content was measured by using ICP-OES Seiko Instruments VISTA-MPX and the dissolved organic carbon (DOC) measurement also carried out by using TOC analyzer Shimazu TOC-V CHS.

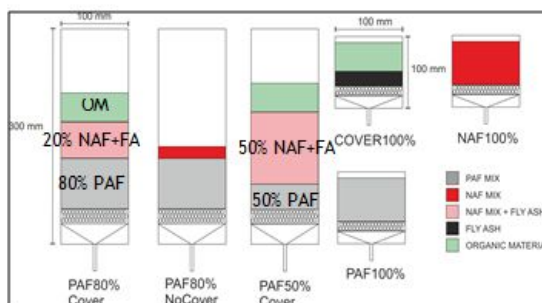


Figure 3: Leaching Column Configuration

Table 2: Weight Distribution of Material in the column

Column	Weight (g)			
	PAF	NAF	FA	OM
PAF80%	800.44	200.44	400.80	150.01
PAF80%NoCover	800.53	200.53	-	-
PAF50%	400.32	400.24	800.6	150.23
PAF100%	600.63	-	-	-
NAF100%	-	600.81	-	-
Cover100%	-	-	150.68	150.07

## RESULTS AND DISCUSSIONS

From the simulation results, leachate water behavior changes were observed. Wetting phase causes the water flowing through the column after having interaction with the material layer. These consist of water-organic material interaction, water-mineral interactions and also mixing of those waters that produce leachate water as the final output. Therefore, the quality of water and the dissolved matter content in the leachate were observed to obtain information about the possible reaction within the column. Meanwhile, drying phase will reduce the moisture content by evaporation and let the mineral to be oxidized further by the oxygen and affects the AMD generation.

### pH and Electrical Conductivity (EC)

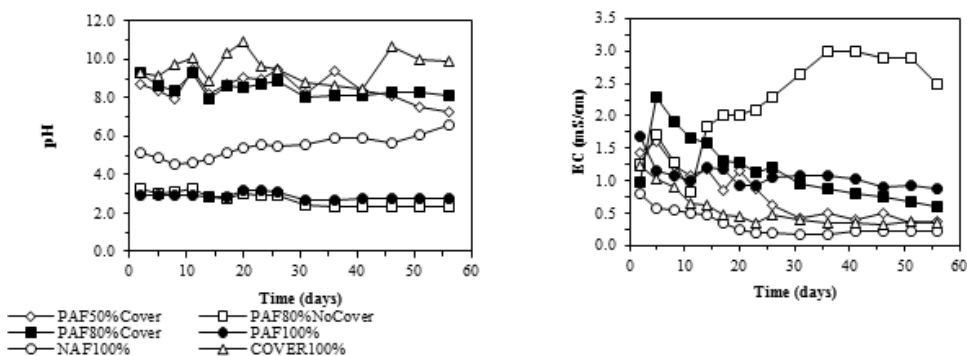
The pH and EC measurement result of 6 columns was presented on the Figure 4. During the simulation, mostly columns had stable pH value. Only NAF100% column that had an increasing pH trend. This possibly happened because the acid generation decreased due to the sulfide mineral was depleted. Compared to the PAF Mix, the sulfur content of NAF Mix was lower with 0.99% difference in Total Sulfur test, which equals to  $MPA=30.30 \text{ kg H}_2\text{SO}_4/\text{ton}$ .

Column PAF100% resulted in the acidic leachate water, with pH value ranged from 2.71–3.21 and the average value was 2.88. This result was expected, since from the mineralogical analysis the PAF material has a high total sulfur concentration and existence of pyrite and arsenopyrite. Moreover, static test has analyzed the acid producing capacity of PAF Mix material which showed high potential in generating AMD. Interestingly, PAF80%NoCover also produced leachate water that in the similar value with PAF100%, even though in PAF80%NoCover column contained NAF layer material above PAF layer. PAF80%NoCover pH value ranged from 2.34–3.31 with the average pH value was 2.75. This

suggests that with the case of the huge difference between PAF volumes available at the mine site, method of layering PAF with NAF layer is ineffective to improve the AMD in the waste dumping area.

On the contrary, the column with the fly ash and organic material cover, PAF50%Cover and PAF80%Cover columns, produced near neutral to alkaline leachate water. PAF50%Cover pH ranged from 7.28–9.48 with the average pH value was 8.52 while PAF80%Cover pH ranged from 7.95–9.30 with the average pH value was 8.47. These results were in concordance with the result of Cover100% column that has a high pH value of leachate water, ranged from 8.48–10.89 with the average 9.59. The alkaline solution from fly ash and organic material cover layer improves the AMD by consuming the acidity. Therefore, even though PAF Mix could generate acidic water, the water from cover material able to buffer pH and improve the water.

Based on the measurement result, PAF80%NoCover had the highest EC on the average (see Table 3), as expected from the low pH measurement result. The greatest dissolved ions concentration can be expected from this column. By comparing the EC from the PAF80% with and without cover material, the significant value difference can be seen. Therefore, it was assumed that the cover had contributed in decreasing the number of dissolved ions of the leachate water. In the case of the lower ratio of PAF material, PAF50%Cover column also showed lower EC value that indicates even less ions dissolved in the column than PAF80%Cover. This result supports the fly ash and organic material contribution assumption previously.



**Figure 4: pH and EC Leachate Water Measurement Result**

Among the columns, the lowest EC value was owned by NAF100% column. Even though the pH of NAF100% was lower than Cover100% column, EC of Cover100% was constantly higher than NAF100%. This possibly happens because the rich oxide content, for example CaO, in the fly ash easily dissolves in the presence of water regardless the pH. In addition, the single layer columns with lower mass (PAF100%, NAF100% and Cover100%) had a lower EC value compared to the column with layer combination. This was understandable since higher mineral concentration in the column means more ions likely to be dissolved in the leachate water. From the investigation, good correlation between the pH and EC trend could be observed. Furthermore, PAF80%NoCover column had the highest EC value on the average, 2.85 mS/cm

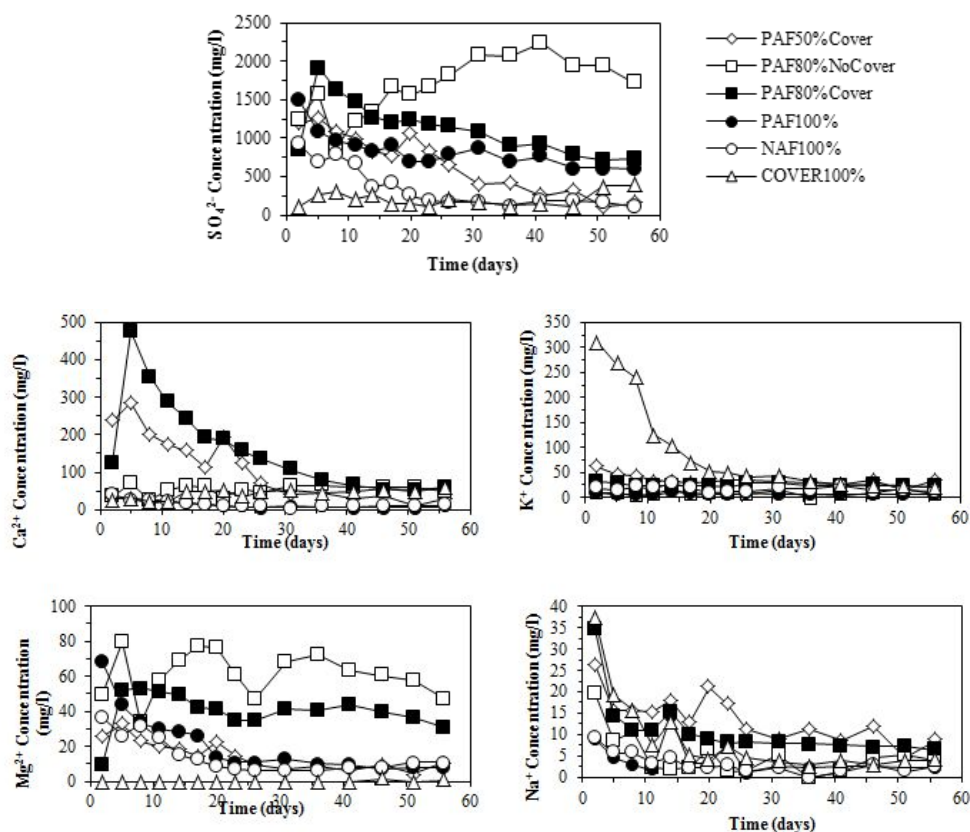
meanwhile the lowest EC value was PAF50%Cover column.

### Major Cation and Anion

Most groundwater contain great concentrations of calcium ( $\text{Ca}^{2+}$ ), magnesium ( $\text{Mg}^{2+}$ ), sodium ( $\text{Na}^+$ ) and potassium ( $\text{K}^+$ ) as major cations and bicarbonate ( $\text{HCO}_3^-$ ), sulfate ( $\text{SO}_4^{2-}$ ) and chloride ( $\text{Cl}^-$ ) as major anions. The dissolution of minerals happens as the main natural sources of cation-anion presence in the water (Younger, 2007). Based on the result of major cations-anions measurement, similar trend could be observed from the initial flush of all columns. All columns showed anomaly initial data that was extremely high. This was expected as shock loading phenomenon, which happened because of the weathering that occurred previously. First flushing caused the readily oxidized mineral to dissolve in the water, thus increased the total major cation-anion. After neglecting the initial measurement, steady changes could be seen that formed certain trend of major cation-anion dissolution.

In the simulation, the major anion was sulfate while the major cations were calcium, magnesium, sodium and potassium. From the result, the largest sulfate concentration was found from PAF80%NoCover column, around 2237.64 mg/l on average. In this column, sulfate anion was likely dissolved from the weathering of sulfide mineral, pyrite and arsenopyrite. In the groundwater system, the principal natural sources of sulfate dissolution are the weathering of sulfide mineral, gypsum and/or anhydrite. Correspondingly, both gypsum and anhydrite could also become the main source of calcium concentration in the groundwater that happens as major cation in the groundwater system. Based on the sulfate measurement (see Figure 5), it can be said that PAF80%NoCover column has the most acidity generated within the column. This was also supported by the lowest pH value among the columns as mentioned earlier. Interestingly, PAF80%Cover had quite a high sulfate concentration in the beginning. However, as the time passed, the sulfate kept decreasing and finally had the similar sulfate concentration like the other columns. This trend was in contrast with PAF80%NoCover column sulfate concentration trend. It indicates the role of cover material in changing the sulfate dissolution behavior.

Generally from major cation anion concentration result, it can be observed different trend depend on the major element of cation in the column. A significant amount of calcium can be seen from the measurement result of both PAF50%Cover and PAF80%Cover, 116.18 mg/l and 172.08 mg/l on the average, respectively. Column without fly ash and organic material, PAF80%NoCover, gave lower value than column with cover, which means a cover layer contributed for the increasing calcium in the leachate water. The calcium content of the column without the cover possibly was produced from silicate mineral dissolution while most calcium of column with cover was originated from fly ash that contain high calcium, in the form of lime ( $\text{CaO}$ ), gypsum and dolomite, based on the XRD and XRF analysis. Those minerals dissolve after reacting with water, that is supported by the pH measurement result which significantly increased. The dissolution of those minerals produces the hydroxide ion which will consume acidity and increase the water pH.



**Figure 5: Major Cation-Anion Measurement Result of Each Column**

Potassium (K) dissolution showed a stable trend for the rest of the column, except for Cover100% column for the initial data. After time passed, the K concentration remained stable. On the contrary, magnesium showed a noticeable difference in measurement for each column. The smaller column, which is PAF100%, NAF100% and Cover100%, gave a higher result of Mg for PAF100% compared to other 2 columns. Mg usually dissolved from carbonate mineral, together with Ca source such as carbonate and silicate minerals. This cation also can come from weathering product of minerals, especially clay minerals. Since the pH of leachate water in PAF80%NoCover was low, the concentration of Mg in this column higher as the dissolution result of acidic water. For sodium (Na), the source of this cation was expected from mineral in fly ash dissolution, since Na was increased in the column with cover rather than without the cover. Moreover, it also confirmed by the higher concentration of Na in the Cover100% rather than in PAF100% and NAF100%.

Based from the measurement result, the column that produced the most balance charge between major cations and anions was COVER100%. This happened because the leachate water was constantly in the high pH, so that the dissolution of heavy metals barely occurred. Only common major cations and anions presence in the leachate water. However, PAF50%Cover and PAF80%Cover had an imbalance portion between cations and anions. All of them had greater sulfate concentration, showed by the data plotting below  $\Sigma\text{cations}=\Sigma\text{anions}$  linear line. There was slightly different between



PAF50%Cover and PAF80%Cover, however, compared to the rest the PAF80%NoCover imbalance was larger. Sulfate concentration dominated the PAF80%Cover, which means in this column there was a larger cations dissolution that came from sulfide oxidation since the sulfate concentration was anomalous. Therefore, it could be expected from the column with the cover to have a high dissolved metal concentration that possibly obtained from sulfide mineral oxidation or else from the dissolution of mineral as chemical weathering products.

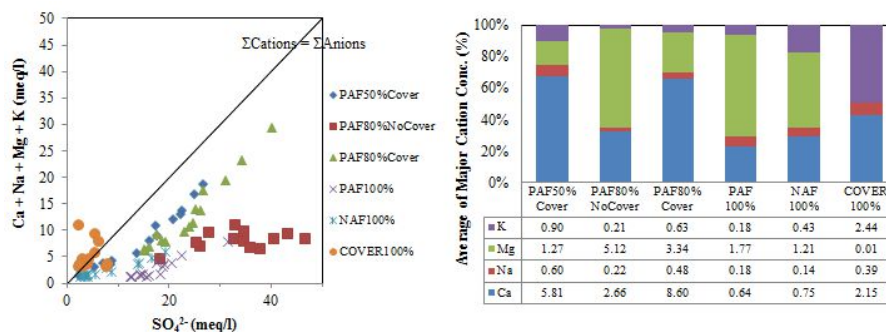


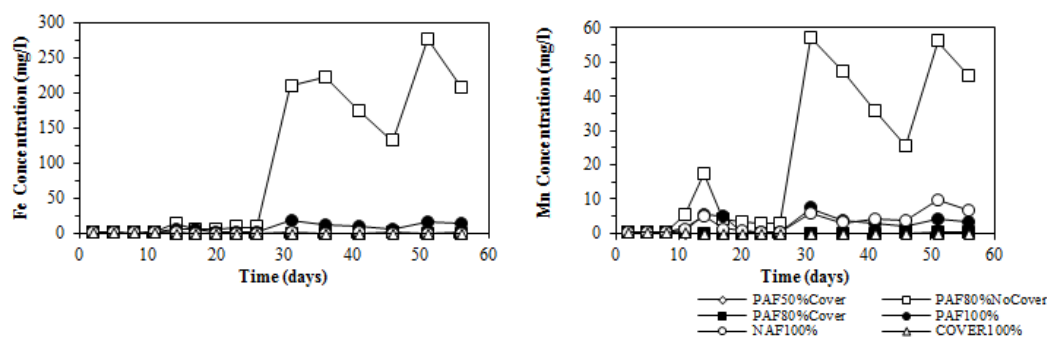
Figure 6: Major Cations and Anions in the Leachate Water of Column

Figure 6 provides the bar chart of major cation concentration portion on average of each column. Using the chart, dominant cation can be seen statistically in each column. PAF50%Cover and PAF80%Cover showed similar portion, where the dominant cation was calcium. PAF100% and NAF100% column each separately had contributed in the calcium concentration in the column based on their major cation that was produced, however the major calcium concentration was obtained from cover material, which assumed came from fly ash. Magnesium likely originated from PAF and NAF material, as shown in the portion of PAF100% and NAF100% column. Cover100% showed an insignificant amount of magnesium, therefore the previous assumption becomes rational.

Similar like Ca, potassium in the leachate water of column with cover material has high possibility had been dissolved from material in the cover. This could be originated from mineral in the fly ash and organic material. This in accordance with the result from the study of composition of EFB by AdeOluwa (2005) that showed the dominant elements were: N (0.97%), C (45.00%), C:N (46.68%) and K (0.97%).

**Dissolved Metal Concentrations**

Measurement by ICP-OES showed the dissolved metal concentration in the leachate water as the result of the wetting by flushing and drying phase. Similar trend could be observed from iron (Fe) and manganese (Mn) metal concentration. The measurement result of metal was provided in the Figure 7. Based on the graph, it can be seen that during the simulation there was a sudden increase of the metal concentration. This happened because during the experiment, the duration of flushing period was extended, from every 2 days to become every 4 days. This was conducted in the consideration of stable pH of leachate water and EC; therefore, longer drying phase to increase the reaction rate was applied to the column. Thus, higher dissolved metal concentration could be seen as the result.

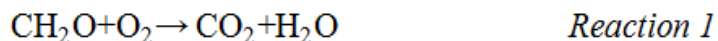


**Figure 7: Metal Concentration Measurement Result**

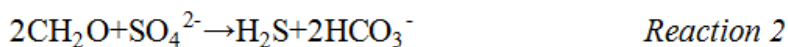
Total iron dissolution concentration on the average was very high for PAF80%NoCover and PAF100% column, 276.54 mg/l and 19.22 mg/l, respectively. The other columns had iron concentration which is lower than 1 mg/l. Even though the trend of iron dissolution based on the measurement result was also similar to Al and Cu trend dissolution, the noticeable dissimilarity can be discovered. PAF80%NoCover and PAF100% column had a greater difference in the value of iron concentration than Al and Cu. This possibly happened because the iron dissolution source had mainly been from sulfide mineral oxidation by the oxygen. This oxidation reaction produces more reaction product as the contact time between surface of sulfide mineral and oxygen in the air is increased. As a consequence, the dissolved iron also increases significantly. For Mn, not only PAF80%NoCover and PAF100% column that had a significant number in the concentration, but NAF100% column also had a slightly higher concentration compared to the other columns. Therefore, in the system of column with and without cover, NAF rocks contributed in increasing the Mn.

#### **Total Dissolved Organic Carbon**

Organic material in the cover layer was obtained from EFB material. EFB contains mostly organic carbon (C) that could be simplified as  $\text{CH}_2\text{O}$ . This layer acts as an active part for reacting with oxygen during its degradation process under aerobic condition, which has reaction as in the reaction 1.



From reaction above, when the organic material decomposition take place it will have a role in consuming the oxygen when it penetrates the layer. This process will reduce the possibility in the AMD generation since initial oxidation of sulfide mineral needs oxygen as the oxidizing agent. Furthermore, when the organic material dissolved in the water and flowing to the PAF layer where sulfate anion is present, both will react together to produce hydrogen sulfide ( $\text{H}_2\text{S}$ ). Then  $\text{H}_2\text{S}$  will bind together with metal to form metal sulfide and precipitate. This reaction could occur in the presence of sulfate reducing bacteria (SRB) which use the organic material as carbon source and electron donor. Sulfate in this process is used as electron acceptors for its energy generation. This reaction only can be carried out on the condition without the presence of oxygen, because SRB is an anaerobic bacteria. This process follows reaction based on the equation 2.



From the reaction 2, it can be assumed that the degradation process of organic material has a function to improve AMD negative effect. Aerobic process could reduce the oxygen level that penetrates the layer while anaerobic process, with the presence of SRB, can consume the sulfate anion, bind the dissolved metal in the solid form and contribute in the alkaline matter content for leachate water.

In the column leaching experiment, the weight of EFB was distributed evenly for the column that has cover layer, as mentioned earlier on the Table 3. Based on dissolved organic carbon (DOC) measurement, column without cover also contain organic carbon because their results always more than zero (see Figure 8).

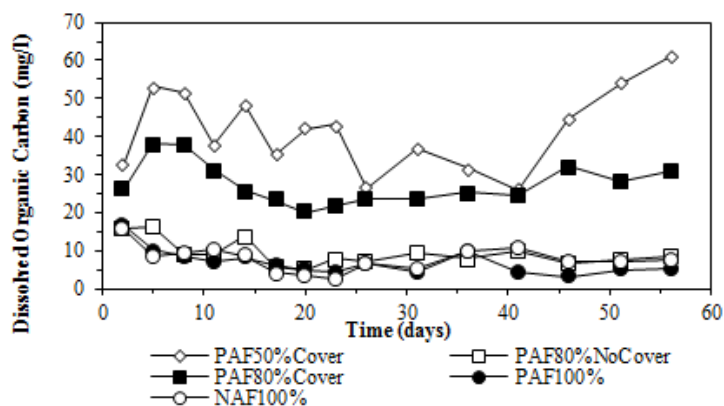


Figure 8: Dissolved Organic Carbon Measurement

NAF100%, PAF100% and PAF80%NoCover showed an insignificant difference in DOC concentration. On the contrary, PAF80%Cover and PAF50%Cover had an amount of DOC higher than the columns without the cover. It implies that organic material had reacted in the cover by aerobic respiration, hence consumed the oxygen. Interestingly, even though both of the covers had the same weight amount of organic material, PAF50%Cover had a constant higher amount of DOC rather than PAF80%Cover. There are two possibilities that might be occurred in the column with cover. Firstly, fly ash has a significant amount of organic material that contributes in the dissolution of organic carbon in the leachate water. Secondly, the dissolved carbon in the water from the cover layer was flowed through the column in similar amounts. Since PAF80%Cover had more sulfate anion, it reacted with the dissolved organic matter in the unoxidized layer of PAF, because during the reaction an environment without oxygen is required. Hence the DOC amount in the PAF80%Cover was lower than PAF50%Cover column.

## CONCLUSIONS

In the case of PAF abundance, conventional method of dry cover system by placing NAF rock to the PAF rock is not efficient for AMD prevention method. Acidic water with dissolved heavy metals is still generated in the similar

behavior with the PAF rock without the addition of NAF rock. Therefore, this method needs to be improved. Utilization of fly ash and organic material as cover layer is effective to prevent AMD generation, by increasing the leachate water pH, consuming the oxygen in the upper layer and reducing the dissolved metal concentration. The dissolution of heavy metals from fly ash seems insignificant in this experiment. Based on the measurement of major cation-anions, dissolution of calcium from fly ash can be seen in the leaching column. Calcium can react with sulfate and formed the gypsum therefore decrease the permeability of the layer. Moreover, the TOC measurement also showed the increasing value of DOC in the column with fly ash and organic material cover, thus possibly react with sulfate and precipitate the metals. Fly ash and organic material combination as cover layer thus far shows no negative impacts may happen due to its utilization. Therefore, it is important to conduct this experiment in the field scale to know the behavior of leachate water in the field situation.

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