Acid Mine Drainage Prevention through the Dry Coating Method Using Fly Ash and Bottom Ash

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ABSTRACT

Acid mine drainage poses a significant challenge in open-pit coal mining, particularly in Indonesia, necessitating the development of effective prevention and control methods. This research aimed to assess the impact of fly ash (FA) and bottom ash (BA) coating on the mixing ratio of potentially acid-forming (PAF) and non-acid-forming (NAF) materials using a leaching column over an 8-week period. The results revealed a trend of increasing pH values and decreasing sulfate and metal concentrations in the leachate. Mixing 25% FABA in both PAF scenarios produced leachate with total concentrations of S, Fe, and Mn at 0.026 mg/L, 0.117 mg/L, and 0.677 mg/L, respectively, with a pH close to neutral (6.28). Kinetic tests indicate TDS and EC values in the resulting leachate at 1221 ppm and 2442 µs/m. This study demonstrated that PAF coating using NAF and the fly ash bottom ash (FABA) is an effective method for preventing or minimizing the generation of acid mine drainage.

Keywords: leachate, leaching column, PAF, NAF, coal mining.

INTRODUCTION

Indonesia stands as the world foremost coal producer. By 2018, the annual coal production had escalated to 548.6 million tonnes, with a mere 114.55 million tonnes allocated for domestic consumption. In accordance with British Petroleum data from 2019, Indonesia cumulative coal resources are estimated at approximately 140.48 billion tons, encompassing 29.91 billion tons identified as coal reserves, thereby constituting 3.5% of the global overall coal reserves. Broadly speaking, coal mining practices in Indonesia predominantly involve open-pit mining techniques. Within this framework, one of the key operational activities revolves around stripping and disposing of overburden materials, a process that carries the inherent potential to yield acid mine drainage. Acid mine drainage is an outcome of the oxidative reaction of sulfide minerals, which are exposed during mining and interact with the surrounding atmosphere [Said, 2018].

In the context of open-pit coal mining, the genesis of acid mine drainage arises from active mine holes and accumulations of overburden material. This process persists until the post-mining phase. Consequently, it is imperative to formulate preemptive strategies to minimize the generation of acid mine drainage in the post-mining stage [Abfertiawan et al., 2020]. The implementation of acid mine drainage mitigation measures commences during the exploration phase, which lays the foundation for mining activities within the pit area. The fundamental approach to preventing acid mine drainage revolves around eliminating one of the primary components involved in the oxidation reaction. These components include

rocks containing sulfide minerals, water (H_2O), and air (O_2) [Miller et al., 2019]. Various methods have been utilized to inhibit the occurrence of acid mine drainage. These encompass the deployment of an oxygen barrier, bactericides, co-disposal and blending, as well as passivation techniques (involving organic coatings, microencapsulation, and silane-based coatings [Moncur et al., 2015; Zhang and Wang, 2017; Fan et al., 2017; Li et al., 2019].

The oxygen barrier technique stands as one of the most widely employed methods. Its primary principle revolves around averting the reaction of rocks containing sulfides (potentially acid forming) with water and air through the utilization of layering. In preventing acid mine drainage, the oxygen barrier encompasses two main methods, namely the dry and the wet cover. The most prevalent method for preventing acid mine drainage is the employment of the dry cover technique, which involves the use of non-acid-forming (NAF) materials to overlay potentially acid-forming (PAF) materials. This approach is preferred because it is effective and efficient [Matsumoto et al., 2017]. Wet covers, on the other hand, need special topographical and hydrological conditions, such as the proximity of lakes or rivers near mining regions. This makes wet coverings less suited for large-scale applications, particularly in seismically active areas where the danger of wastewater leakage is enhanced.

Several studies have been conducted to investigate the techniques for preventing acid mine drainage using the dry cover approach. Non-Acid-Forming (NAF) materials are used to cover Potentially Acid-Forming (PAF) materials [Abfertiawan et al., 2020; Roychowdhury et al., 2015], lime treatment [Zhou et al., 2017], cement application [Sephton and Webb, 2019], and the combination of portland cement with fly ash [Sephton et al., 2019]. Additional research has looked into the usefulness of various alkaline materials, such as fly ash and bottom ash, as PAF cover materials. These materials have been shown to be efficient in lowering metal concentrations, such as Fe and Cu, in acid mine drainage [Nfissi et al., 2017; Win et al., 2020]. As far as it is known, the research focusing on avoiding acid mine drainage development utilizing the dry cover material approach and simulated by leaching columns is limited. Consequently, this research aimed to evaluate the impact of using fly ash and bottom ash materials in different ratios of PAF and NAF materials over an 8-week period. The study also involves the analysis and comparison of metal content and leachate characteristics produced during the kinetic testing.
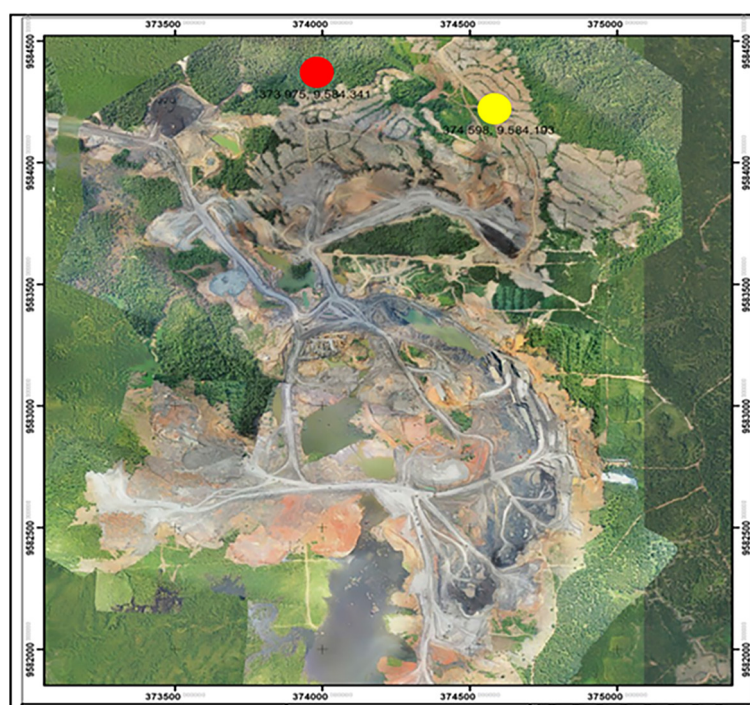

Figure 1. The location of PAF and NAF sampling

MATERIALS AND METHODS

Materials

Materials (PAF and NAF) were obtained from the mining area located at PT. Bukit Asam in the Muara Tiga Besar, Tanjung Enim, South Sumatra, Indonesia. Two claystone samples were collected, one with the potential for acid formation, referred to as PAF material, and the other incapable of forming acid, referred to as NAF material. The samples were selected from distinct lithologies. The PAF samples, totaling 30 kg, were sourced from the B2–C interburden layer, classifying under the light PAF category on the north side of the Muara Tiga Besar Location (N 364757, E 9588563). On the other hand, the NAF sample, also weighing 30 kg, was extracted from the interburden A1–A2 on the south side of the Muara Tiga Besar location (N 364411, E 9584865) (Figure 1). All samples underwent crushing using a jaw crusher to achieve dimensions smaller than 2 cm. The fly ash and bottom ash were obtained from a coal steam power plant located in the coal mining area of PT. Bukit Asam.

Static and mineralogical test

Static tests were carried out to evaluate the AMD generation capacity, encompassing examinations of paste pH, acid-base account (ABA), total sulfur, and net acid generation (NAG). The minerals in the materials were assessed thru mineralogical analysis using X-ray fluorescence

Table 1. Materials mixture leaching column ratio

Sample	Description
A	NAF 100%
B	PAF 100%
C	PAF 50% + NAF 50%
D	PAF 50% + NAF 25% + FA 12.5% + BA 12.5%
E	PAF 50% + FA 25% + BA 25%

(XRF). The material morphology was analyzed using scanning electron microscopy (SEM).

Leaching column test

The leaching column experiment was performed utilizing a transparent acrylic column with a 15 cm diameter and a 30 cm height, as depicted in Figure 2. The sample height measured from the bottom of the filter, was maintained at 25 cm. Porous beds were placed at the boundary areas corresponding to the material mixing ratio. On the upper part of the column, a 60-W lamp was positioned to replicate sunlight and uphold an average temperature ranging from 30 to 35 °C, reflecting the conditions prevalent in the South Sumatra coal mining study area. Every week, 1 liter of deionized water was added to each sample column to simulate wet or rainy cycles. The infiltrated water into the rock sample streamed beneath the column and was gathered for subsequent analysis. The simulation included columns with various thickness ratios of FABA mixed with PAF rock, as outlined in Table 1. This approach was employed to investigate the cover impact on
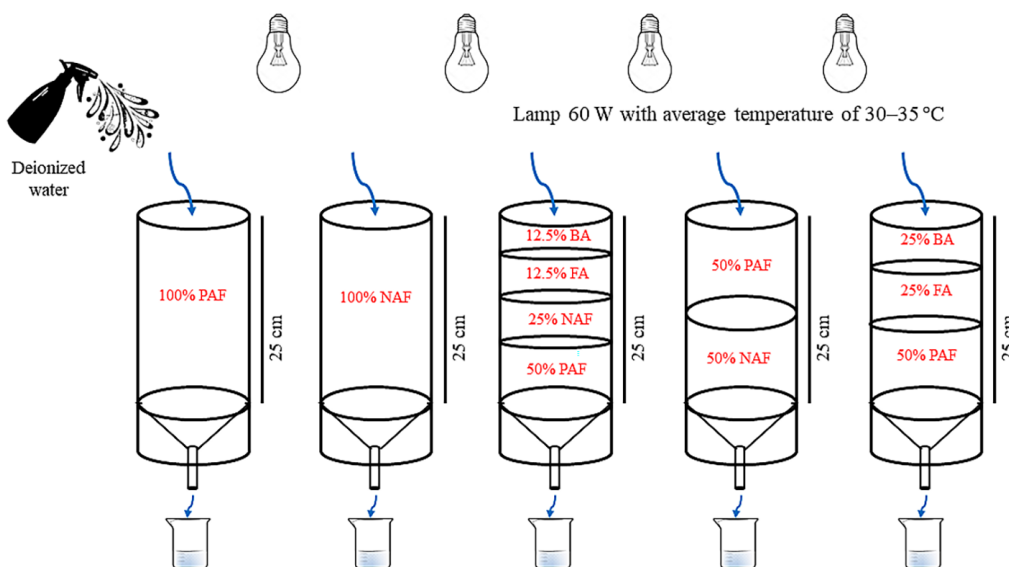

Figure 2. Leaching column configuration

AMD generation as well as to compare the effectiveness of different fly ash and bottom ash ratios within the cover.

The leaching test spanned a total duration of 8 weeks, involving wetting and drying phases designed to mimic the conditions in mining fields. During the drying stage, a daily cycle was established, consisting of 12 hours of lamp heating and 12 hours of natural drying. For the wetting phase, deionized water spraying was implemented, and the leachate volume was maintained at 150 mL per cycle. Subsequent to leachate emergence, the pH and electrical conductivity (EC) were promptly measured. The pH measurements were conducted at each layer (boundary plane) of the material mixing ratio. The leachate was then subjected to filtration using a 0.45 μm filter and subsequently acidified. Cation-anion measurements were performed employing a Dionex ICS-90 ion chromatograph.

RESULTS AND DISCUSSION

Identification of acid mine water through static test

The outcomes of the static examinations indicate that the sample categorized as potentially acid-forming (PAF) exhibits an acid-neutralizing capacity (ANC) of 31.59 kg H_2SO_4/ton. This measurement quantifies the volume of neutralizing material, encompassing carbonate compounds within the rock, accessible for the neutralization of acid. The elevated acidity is also reflected in a low net acid generation (NAG) pH of 2.62, accompanied by a positive net acid-producing potential (NAPP) value of 43.63 kg H_2SO_4/ton. NAPP represents the equilibrium between maximum potential acidity (MPA) and ANC. A positive NAPP value indicates the potential for acid formation.

The PAF sample had a relatively high total sulfur (S) percentage of 2.46%. The total S analytical approach employed in this work is considered conservative, since it does not distinguish between different S forms in the sample, such as

sulfide and sulfate. It is important to note that native sulfate and S are not considered contributors to acid mine drainage. The static test outcomes, depicting the attributes of the two samples (PAF and NAF), are presented in Table 2. The first sample, S-1, falls under the PAF category, while S-2 is classified as NAF. Sample S-1 exhibits a greater acid formation potential than sample S-2, with a maximum potential acidity (MPA) value reaching 75.22 kg H_2SO_4/ton, justifying its classification as PAF.

Characteristics and mineralogical test of materials

Mineralogical and morphological analyses were performed on the soil samples obtained from former mining areas and coal ash to identify and verify the varieties of sulfide minerals existing in each sample. Figure 3 illustrates the SEM and XRD results for each sample. The surface morphology of the potentially acid-forming (PAF) sample is notably dense and smooth, exhibiting a slightly porous surface, while the non-acid-forming (NAF) sample displays a less regular surface. The fly ash bottom ash (FABA) material demonstrates a large porous surface morphology with cavities. Sample S-2 can be categorized as NAF, since it comprises minimal minerals containing sulfur but includes carbonate minerals, both of which contribute to high acid-neutralizing capacity (ANC) concentrations. The dominant minerals observed in PAF include SiO_2, Al_2O_3, Fe_2O_3, and SO_3. Framboidal pyrite minerals in sample S-01 act as the primary source for acid mine drainage formation. The morphological features suggest that pyrite minerals may undergo a more rapid oxidation process due to their more surface area. Conversely, the prevailing minerals in the NAF include Al_2O_3, SiO_2, and CaO. The presence of sulfate mineral in the PAF samples has the ability to create sulfate and H ions, contributing to the formation of acid mine drainage along river flows [Yang et al., 2023].

From Table 3, the concentration of Fe_2O_3 in the PAF material is notable, suggesting the

Table 2. Static test result

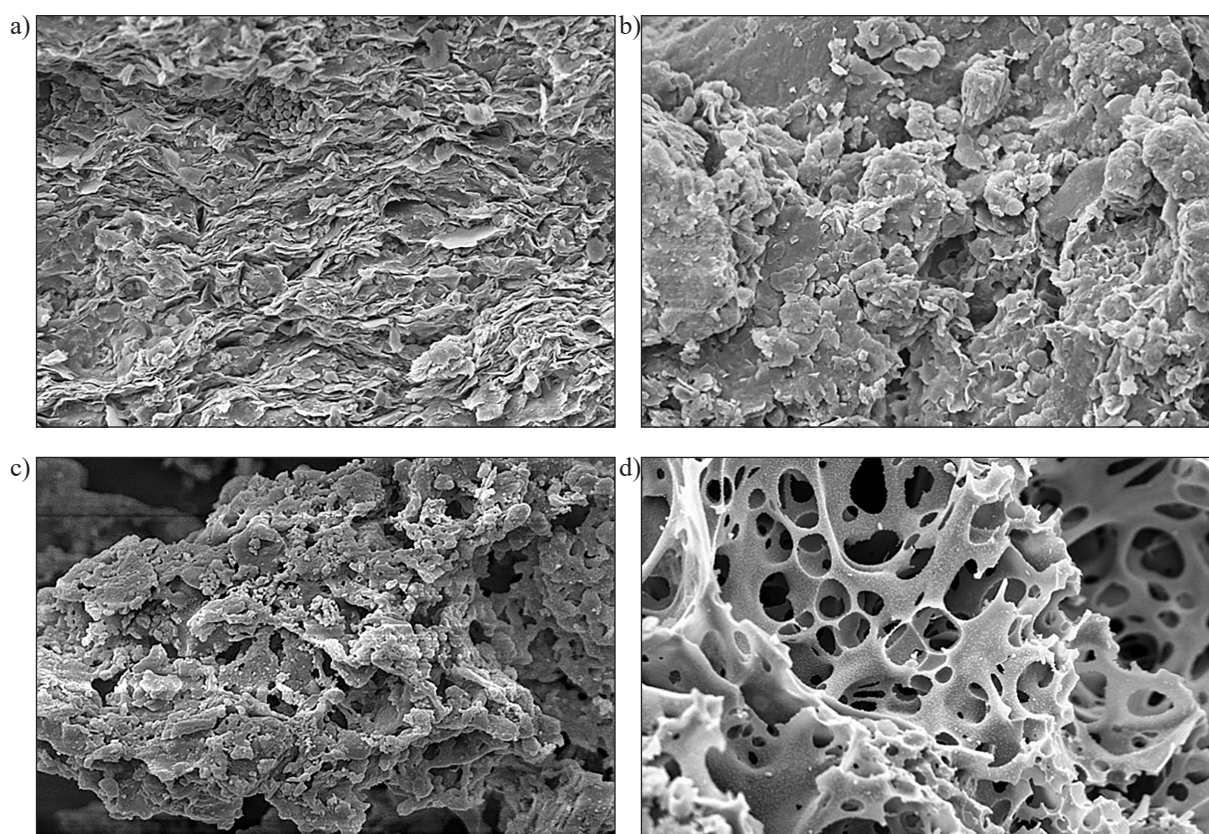| Sample	Total sulfur (%)	MPA	ANC	ANC/MPA	NAPP	pH NAG	NAG		Category
							pH 4.5	pH 7	
S-1	2.46	75.22	31.59	0.42	43.63	2.62	24.82	30.49	PAF
S-2	0.77	23.51	29.63	1.26	-6.12	8.18	0	0	NAF

Figure 3. Morphology characteristic of (a) PAF, (b) NAF, (c) fly ash, and (d) bottom ash

Table 3. Composition (%) of oxide chemical compounds from each material

Component	PAF	NAF	FA	BA
MgO	0.852	0.852	1.302	0.439
Al_2O_3	25.273	13.973	23.977	22.391
SiO_2	56.362	62.362	50.429	55.223
P_2O_5	1.194	1.194	2.994	4.194
SO_3	6.374	0.869	0.000	0.000
K_2O	1.890	1.417	1.862	2.329
CaO	0.873	16.286	14.874	10.203
TiO_2	0.897	0.506	1.773	1.719
Fe_2O_3	6.114	2.345	2.332	3.410
Others	0.171	0.196	0.457	0.092

potential production of FeS_2 or pyrite compounds. Sulfide compounds from the PAF material contribute to the overall sulfur content, mainly in the form of SO_3. In contrast, the NAF material contains higher levels of CaO with lower SO_3 concentrations. Fly ash and bottom ash (FABA), designated as cover materials to support NAF, do not contain significant amounts of SO_3, although Fe_2O_3 is still present. Additionally, the CaO composition is comparable to that of NAF, making FABA a suitable cover material to be used in conjunction with NAF.

Leachate characteristics (pH, TDS, and EC)

Simulation tests for PAF layering, utilizing FABA, were conducted to assess their effectiveness and the influencing factors in preventing the formation of acid mine drainage. Leaching column experiments were conducted for a duration of 8 weeks, during which the properties of the leachate from different column conditions were examined to understand the dynamics of acid mine drainage formation and the effectiveness of the NAF and FABA materials. The leach column pH test results are illustrated in Figure

4. The simulation results for the column having 100% PAF and NAF material correspond to the static test results. The leachate from the NAF sample column exhibited a pH of 9, indicating a dominance of alkali carbonate compounds. This aligns with the static test results for sample S-01, revealing a positive acid-neutralizing capacity of 29.63 kg H_2SO_4/ton and an NAG pH of 8.18. The presence of vaterite ($CaCO_3$) and siderite ($FeCO_3$), carbonate minerals, is responsible for the pH characteristics of the leachate. In contrast, the PAF samples exhibit more acidic characteristics. The pH value of leachate from PAF ranges between 2.62–3.65, indicating the formation of acid mine drainage with a pH <4, leading to increased metal solubility. The static test results further support the difference in the potential for acid mine drainage formation between the two materials. The mineral characteristics contribute to the NAF leachate having relatively non-acidic properties compared to the PAF leachate.

On the basis of the analysis results, a low initial pH indicates an initial release of significant amounts of stored acidity. It is concluded that the PAF material has become acidic with a high stored acidity content, implying the continuous production of acidic waste. The pH value in the C ratio (50% PAF and 50% NAF) significantly increased from 3.46 to 5.46 from the first week to the eighth week with the addition of NAF. The most substantial increase in pH occurred after the eighth week of the leaching process, with the addition of 25% NAF, 12.5% FA, and 12.5% BA. According to the outcomes of the kinetic test, the proportion of incorporating fly ash and bottom ash influences the pH value. The presence

of fly ash material proves effective in hindering the oxidation of the PAF layer, as indicated by the pH value of the water infiltrated into the layering column, which remains in proximity to a neutral pH value. The slight increase in the leachate pH observed from 5.48 in the sixth week to pH 6.28 in the eighth week may indicate an increase in the dissolution rate of FABA in PAF over time. This scenario suggests a gradual increase in pH towards neutral values, also indicating decreased total sulfate, Fe, and Mn concentrations.

The results of backfilling the TDS of leachate are presented in Figure 5. Total Dissolved Solids (TDS) signifies the concentration of dissolved solids in the leachate and correlates with the quantity of dissolved metals. The highest TDS value was observed in the leachate from the 100% PAF column. Meanwhile, the lowest value was obtained by mixing 25% FA and 25% BA in PAF after eight weeks of the simulation process. The lowest TDS value in the eighth week was observed to have a value almost the same as the TDS value in (NAF). This is influenced by the layering of fly ash and bottom ash. The TDS value correlateed with the pH value. A lower pH value is associated with a higher measured TDS value. H^+ ions dissolve metals in leachate under acidic circumstances, increasing pH and electrical conductivity [Alegbe et al., 2019]. If the TDS value is high, the EC value will increase due to the presence of dissolved metals that can conduct electricity [Smith et al., 2022].

The ability of water to conduct electric current, expressed in µS/m, is referred to as electrical conductivity (EC). On the basis of the measurement results, the PAF control column had the highest EC, as expected from the low pH measurement results (Figure 6). The elution of metal
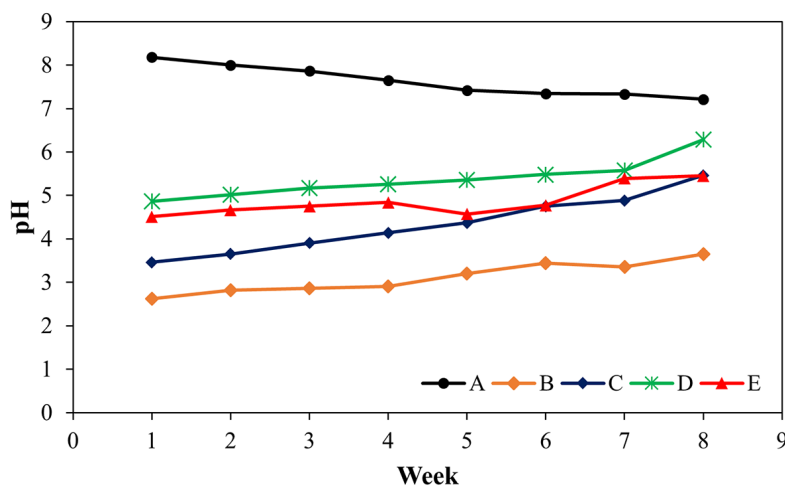

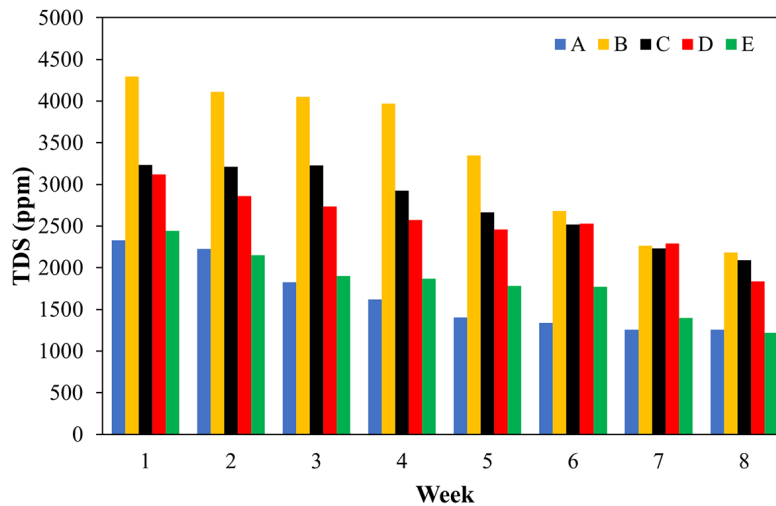**Figure 4.** Changes in the pH of leachate from each leaching column every week

Figure 5. Changes in TDS of leachate from each leaching column every week

ions in an acidic environment occurs because the lower the pH, the more metal ions will elute. Meanwhile, the elution of metal ions is attributed to the neutralization of acids, perhaps partly due to the presence of carbonate minerals, which have a neutralizing effect on acidic water. The electrical conductivity of leachate is very high, about two times higher than with the addition of FABA. The leachate electrical conductivity decreased on average by 51%, from 8596 µS/m to 4368 µS/m for pure PAF, and 54%, from 4656 µS/m to 2512 µS/m for NAF. Therefore, the next column experiment investigated the effect of the system configuration of the PAF leachate treatment system with the help of fly ash and coal bottom ash mixed in various ratios. The higher electrical conductivity of leachate from PAF indicates a higher proportion of dissolved salts caused by the dissolution of metals and associated anions under highly acidic

conditions. Electrical conductivity decreases with the length of the leaching process and the addition of FABA, indicating a low concentration of dissolved salts, possibly due to the capacity of FABA to stabilize and bind metals and other solutes.

Effect of layering on heavy metal and sulfate concentrations

The metal concentration in the leaching process decreased significantly at a FABA mixture ratio of 25%, indicating an increased immobilization effect of fly ash and bottom ash on metals, especially under the conditions close to neutral and alkaline pH. The contribution of increasing pH to metal immobilization and the accompanying decrease in solutes is supported by the significant inverse relationship between pH and EC, and pH and metal concentration. Fly ash and bottom ash
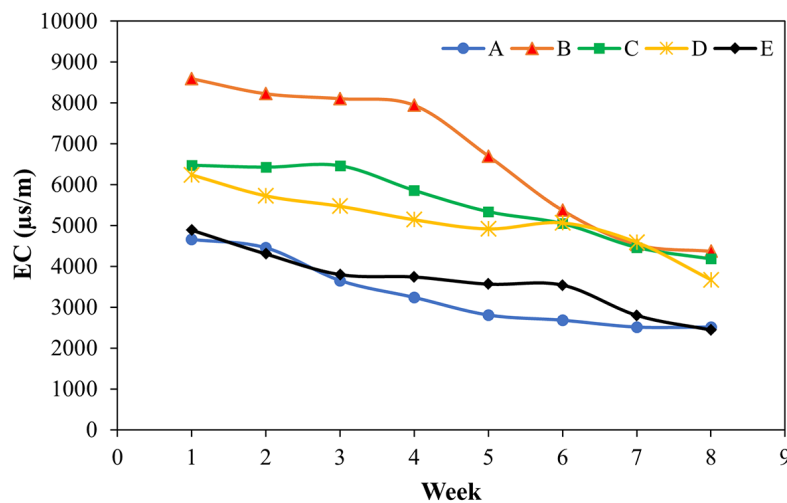

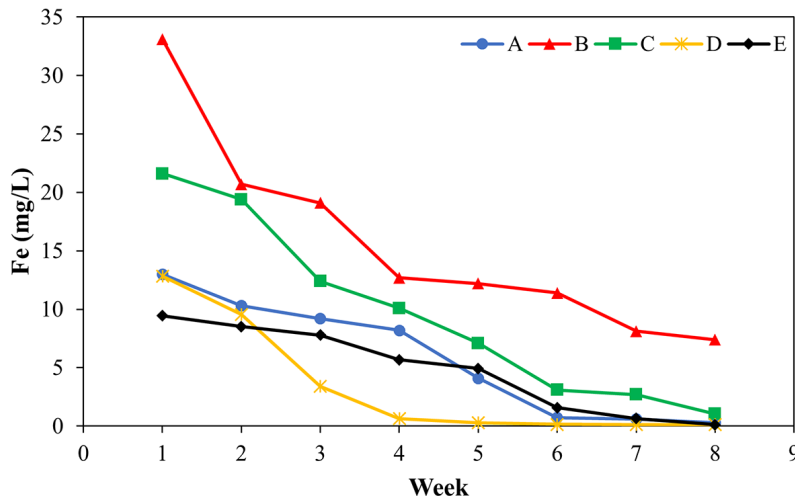**Figure 6.** Changes in the electrical conductivity of leachate from each leaching column every week

Figure 7. Results of total Fe measurements in leachate water

exhibit a high capacity to remove metals through adsorption via ion exchange and surface complexation, even in acidic solutions [Gwenzi et al., 2017; Orakwue et al., 2016].

The total Fe concentration in PAF and NAF leachate was 33.1 mg/L and 13 mg/L, respectively, in the first week. Until week 8, these two elements showed a substantial decreasing tendency (Figure 7), reaching 0.303 mg/L and 7.39 mg/L. When adding 50% NAF in the first week, the total Fe decreased to 21.6 mg/L. The decreasing trend continued until the eighth week (1.04 mg/L). By adding 25% FABA to PAF, by the eighth week, the total Fe had been reduced to 0.117 mg/L. The high Fe content in acid mine water can decrease drastically due to the effects of acid neutralization and oxidation of subsurface water. The addition of FABA has an impact on the absorption of Fe by SiO_2 and Al_2O_3, which have a strong affinity

for electrons [Nidheesh et al., 2012; Orakwue et al., 2016]. SiO_2 and Al_2O_3 in fly ash and bottom ash were found to be 50–55%, and 22–23%, respectively, which is much higher than in NAF. Additionally, the Fe_2O_3 value in FABA is relatively low. Therefore, adding FABA enhances the effectiveness of NAF in reducing the formation of total Fe. The addition of NAF and FABA began to meet the quality standards for total Fe values starting from the sixth week to the eighth week.

The initial concentration of total Mn in the first week in PAF and NAF was 2.260 mg/L and 0.907 mg/L, respectively. The total Mn formed during the leaching process up to the eighth week with the addition of FABA is presented in Figure 8. The leaching simulation process shows a significant decrease, especially in the PAF material itself, particularly from the fourth to the eighth week. Through the contribution of NAF
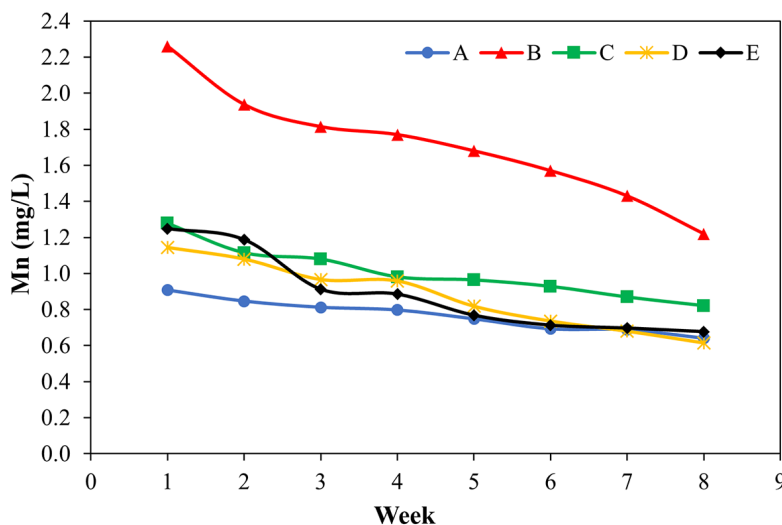

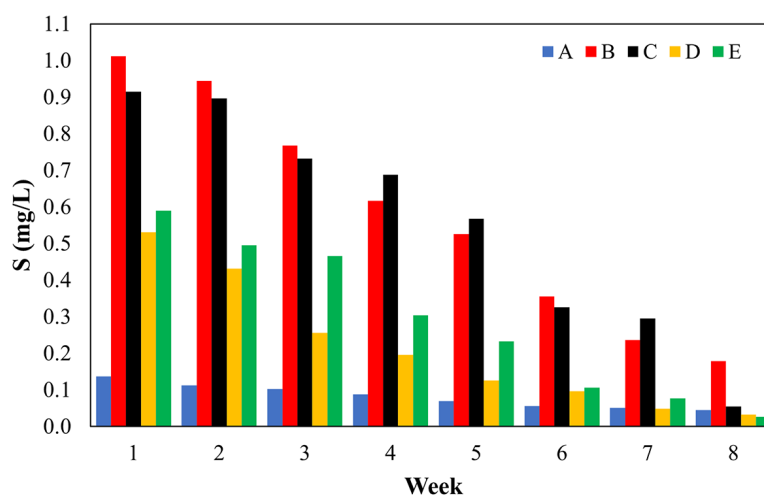**Figure 8.** Results of total Mn measurements in leachate water

Figure 9. Results of total S measurements in leachate water

and FABA in leaching column D, the total Mn was successfully reduced to 0.615 mg/L. Manganese tends to be leached in each leaching column. Manganese solubility is higher in slightly acidic or neutral water, as indicated by the leachate pH, which is close to neutral (6.28). Treating manganese (Mn) in acid mine drainage poses challenges due to its intricate chemistry and the need for a high pH value for precipitation. The research on manganese removal from acid mine drainage using FABA has recognized intraparticle diffusion as the primary rate-limiting step, with additional contribution from boundary layer diffusion when employing small FABA particles [Smiciklas et al., 2023].

Figure 9 illustrates the total sulfate concentration in the leachate from the leaching column. The study findings reveal minimal acid formation in the NAF samples, evidenced by the sulfate concentration in the kinetic test ranging from 1.012 to 0.044 mg/L, averaging 0.288 mg/L over the 8-week testing period (Figure 8). This observation is closely related to changes in the pH value and sulfate content. The addition of 25% FABA succeeded in reducing total sulfate from 0.589 to 0.026 mg/L until the eighth week. The decline in sulfide mineral mass, attributed to reductions in sulfate and metal concentrations in leachate water, resulted in a decrease in the oxidation of sulfide minerals. A 50% reduction occurred with the help of FABA after previously only being able to decrease to 0.054 mg/L using only 50% NAF. Sulfate is the primary oxidation product of prolonged atmospheric exposure, mostly in the form of Fe-sulfate ($Fe_2(SO_4)_3$). $Fe_2(SO_4)_3$ easily dissolves into Fe^{3+} and $SO4^{2-}$ upon contact with

water. Another indication of the occurrence of oxidation in the leachate is the difference in the mass of S before and after the experiment, showing a significant leaching of S (Figure 8). The overall decrease in S during the experiment indicates an ongoing oxidation process.

CONCLUSIONS

The control of acid mine drainage using the layering method in the leaching column was successfully implemented. The results of this research demonstrate that the use of NAF material combined with fly ash (FA) and bottom ash (BA) increases pH as well as reduces sulfate and metal concentrations. According to the kinetic test results, the material mixture consisting of 50% PAF, 25% FA, and 25% BA showed the lowest metal concentration, with total S, total Fe, and total Mn values of 0.026 mg/L, 0.117 mg/L, and 0.677 mg/L. The highest pH of leachate produced until the end of the leaching process was 6.28 in the column with a mixture of 50% PAF, 25% NAF, 12.5% FA, and 12.5% BA. The total dissolved solid (TDS) and electrical conductivity (EC) values in the leachate water were the lowest, namely 1221 ppm and 2442 µs/m. The use of NAF and FABA has proven effective in neutralizing acidity and reducing the concentration of metals available at low cost.

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