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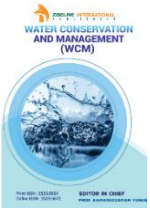

RESEARCH ARTICLE

PASSIVE TREATMENT OF ACID MINE DRAINAGE (AMD) USING AN OPEN LIMESTONE CHANNEL SYSTEM PRIOR TO DISCHARGE INTO SURFACE WATER BODIES

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ABSTRACT

Acid Mine Drainage (AMD) generated from coal mining activities typically exhibits low pH levels (3.75) and high metal concentrations, posing significant threats to aquatic ecosystems. Conventional treatment methods are often costly and less effective in the long term. This study investigates the application of a passive open limestone channel system, utilizing local materials (fly ash, zeolite, limestone, and silica sand) to neutralize AMD. The experimental setup employed three treatment combinations (A, B, and C) tested in an 8-compartment channel system with varying slopes (5°, 7°, and 9°). Combination A used fresh materials, B reused materials from A, and C utilized unwashed materials from B. Effectiveness was evaluated based on pH improvement and operational efficiency. Results showed that Combination A achieved an average pH of 7.51 (76% efficiency), with optimal neutralization occurring in compartment 8. Combination B reached a pH of 7.39 (62% efficiency) but neutralized AMD more rapidly in compartments 5–6. Combination C achieved a pH of 7.15 (73% efficiency). The pH decline in the final compartments was attributed to CO_2 accumulation, gypsum residue formation, and material clogging. Although Combination A had the highest chemical efficiency, Combination B was deemed the most technically optimal due to its balanced pH improvement (7.39), faster neutralization rate, and material efficiency. The study recommends integrating chemical and technical aspects in AMD treatment system design to ensure environmental and economic sustainability.

KEYWORDS

Acid Mine Drainage, pH neutralization, open limestone channel, fly ash, operational efficiency

1. INTRODUCTION

One of the major environmental impacts of mining activities is the formation of Acid Mine Drainage (AMD), a highly acidic water flow with elevated concentrations of dissolved metals. This phenomenon is predominantly associated with coal mining processes (Xu et al., 2022; Anekwe & Yusuf, 2023). AMD is generated through the oxidation of sulfide minerals commonly found in mining sites, such as pyrite and marcasite (FeS_2), pyrrhotite (Fe_xS_x), chalcocite (Cu_2S), covellite (CuS), molybdenite (MoS_2), chalcopyrite ($CuFeS_2$), galena (PbS), sphalerite (ZnS), and arsenopyrite (FeAsS). These minerals react with water (H_2O), oxygen (O_2), and other environmental factors, leading to severe ecological consequences, both directly and indirectly (Fosu et al., 2020; Roulia et al., 2022; Bharat and Gehendra, 2020; Akhyar et al., 2023).

Beyond its environmental harm, AMD presents significant operational challenges for the mining industry, as it must be treated prior to discharge to meet environmental standards. AMD treatment is financially burdensome, yielding no immediate economic return, yet it is a critical indicator of environmental responsibility. Although conventional treatment techniques – such as pH neutralization, iron (Fe) and manganese (Mn) reduction, and control of total suspended solids (TSS) – are regulated by effluent quality standards (Ricardo et al., 2024; Lei et al., 2024; Lei et al., 2023; Hu et al., 2024), their long-term effectiveness remains limited. Contributing factors include natural attenuation processes (the reduction of contaminants through physical, chemical, and biological mechanisms) and the possibility of increasingly stringent environmental regulations in the future (Evgenia et al., 2015; Hedin, 2024). In the process of AMD formation, Stumm & Morgan (1996) proposed a model for pyrite oxidation, as illustrated in Figure 1.

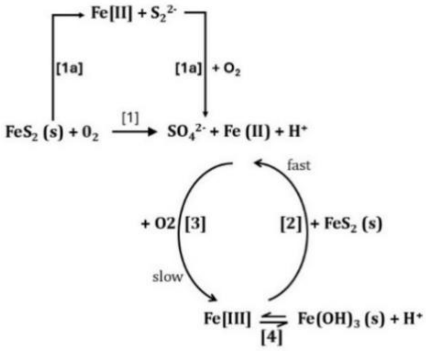

Figure 1: Model for the Oxidation of Pyrite (Stumm & Morgan, 1996)

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The first reaction involves the weathering of pyrite, a sulfide mineral, accompanied by oxidation that produces sulfate and releases ferrous iron. This reaction generates two moles of acidity per mole of oxidized pyrite. Although dissolved oxygen can also participate in the oxidation, its role is less significant due to its limited solubility. This reaction can be written according to the Reaction (1).

$$\underset{pyrite}{2FeS_{2\,(s)}} + \underset{oxygen}{7O_{2\,(g)}} + \underset{water}{2H_2O_{(aq)}} \longrightarrow \underset{ferrous\ iron}{2Fe^{2+}{}_{(aq)}} + \underset{sulfate}{4SO_4^{2+}{}_{(aq)}} + \underset{acidity}{4H^+{}_{(aq)}} \quad (1)$$

Reaction (1) can occur under both abiotic and biotic conditions. In addition to direct oxidation, pyrite may dissolve and subsequently undergo oxidation, where aqueous ferric ions can also act as oxidizing agents for pyrite (Stumm & Morgan, 1996; Youzheng et al., 2020; Lei et al., 2024). Initially, pyrite oxidation begins with Reaction (1) under near-neutral pH conditions, but as acidity increases (pH < 4.5), the process is further accelerated by a secondary reaction pathway (Sandisiwe et al., 2021; Casey et al., 2022).

In Reaction (2), pyrite undergoes further oxidation through interaction with ferric iron (Fe^{3+}), a process that occurs two to three times faster than oxidation by oxygen alone (Stumm & Morgan, 1996; Hao et al., 2021). This reaction generates a significantly higher amount of acidity per mole of pyrite oxidized. The reaction is represented as follows:

$$\underset{pyrite}{FeS_{2\,(s)}} + \underset{ferric\ iron}{14Fe^{3+}{}_{(aq)}} + \underset{water}{8H_2O_{(aq)}} \longrightarrow \underset{ferrous\ iron}{15Fe^{2+}{}_{(aq)}} + \underset{sulfate}{2SO_4^{2-}{}_{(aq)}} + \underset{acidity}{16H^+\,(aq)} \quad (2)$$

In Reaction (2), ferrous iron (Fe^{2+}) is oxidized to ferric iron (Fe^{3+}), consuming one mole of acidity in the process. This reaction proceeds slowly under abiotic conditions at pH levels below 5 (Stumm & Morgan, 1996; Gurkiran et al., 2018). However, the presence of the bacterium Acidithiobacillus ferrooxidans can accelerate this oxidation by a factor of five to six (Stumm & Morgan, 1996; Youzheng et al., 2020). The reaction is represented by Reaction (3).

$$\underset{ferrous\ iron}{4Fe^{2+}{}_{(aq)}} + \underset{oxygen}{O_{2(g)}} + \underset{acidity}{4H^+{}_{(aq)}} \longrightarrow \underset{ferric\ iron}{4Fe^{3+}{}_{(aq)}} + \underset{water}{2H_2O_{(aq)}} \quad (3)$$

The assumption that ferric ions alone can oxidize pyrite in the absence of oxygen is inaccurate. Reaction (3) clearly demonstrates that oxygen is essential for the oxidation of ferrous to ferric iron (Casey et al., 2022; Lei et al., 2024; Kleinmann & Ackman, 2024).

In the fourth reaction, ferric ions produced in Reaction (1) undergo further oxidation and hydrolysis to form ferric hydroxide. This stage involves the precipitation of ferric hydroxide, a process that is highly dependent on pH, occurring more extensively at pH values above 3.5 (Evgenia et al., 2015; Hao et al., 2021). The model of this reaction is presented in Reaction (4).

$$\underset{ferrous\ iron}{Fe^{2+}{}_{(aq)}} + ½O_{2(g)} + 5/2H_{2(aq)} \longrightarrow \underset{\substack{ferric\ hydroxide\\ precipitate}}{4Fe(OH)_{3(s)}\!\downarrow} + 2H^+{}_{(aq)} \quad (4)$$

In the fifth reaction, the combination of Reactions (1) and (4) results in the formation of ferric hydroxide precipitate, commonly known as “Yellow Boy” (Stumm & Morgan, 1996; Gurkiran et al., 2018; Sandisiwe et al., 2021). The reaction is represented by Equation (5), and visual examples along with its treatment approach can be seen in Figures 2 and 3.

$$\underset{pyrite}{FeS_{2(s)}} + \underset{oxygen}{\tfrac{15}{4}O_{2(g)}} + \underset{water}{\tfrac{7}{2}H_2O_{(aq)}} \longrightarrow \underset{\mathbf{yellow\ boy}}{Fe(OH)_{3(s)}\!\downarrow} + \underset{sulfuric\ acid}{2SO_4^{2-}{}_{(aq)}} + 4H^+{}_{(aq)} \quad (5)$$

Regarding ferric ion solubility, it is observed that at pH 4, ferric hydroxide ($Fe(OH)_3$) precipitate begins to form alongside the presence of dissolved Fe(OH)4- species (Hao et al., 2021; Sandisiwe et al., 2021; Casey et al., 2022). The formation of $Fe(OH)_3$ precipitate increases significantly as the pH decreases. At pH levels below 4, iron primarily exists in the dissolved form of Fe(OH)2+. Conversely, at pH values above 12, the formation of $Fe(OH)_3$ precipitate decreases, and the Fe(OH)4- species becomes more dominant. The predominance of $Fe(OH)_3$ precipitate within the pH range of 4–12 plays a crucial role in promoting coagulation processes (Kouhi, 2024). The optimal pH range for minimal solubility, and thus the most effective treatment outcome, is between pH 6 and 8, with the lowest solubility observed at pH 7 (Casey et al., 2022; Lei et al., 2024).

Figure 2: Example of the appearance of a "Yellow Boy" in a mining area.

Figure 3: Example of AMD handling process in a sedimentation pond with neutralizing material, followed by flow through an open channel into a swamp as a surface water reserve.

Based on the above analysis, it is essential to neutralize AMD before it is discharged into surrounding surface water bodies near mining areas. This is because AMD can significantly damage aquatic ecosystems, including rivers, lakes, swamps, and groundwater. Once AMD reaches the stage of forming ferric hydroxide precipitate ($Fe(OH)_3$), commonly referred to as Yellow Boy, the continuation of AMD generation with low pH becomes difficult to halt. This is due to the ongoing nature of the reaction, which persists until one of the reactants is completely depleted (Stumm and Morgan, 1996; Youzheng et al., 2020; Lei et al., 2024).

Several previous studies have identified materials capable of neutralizing AMD pH. Rusli & Samosir (2021) utilized fly ash and bottom ash (FABA) to neutralize AMD, showing that the application of 1 gram of FABA in 200 mL of AMD could increase the pH from 3.72 to 7.30. Nurfasiha & Kusuma (2020) simulated AMD management using an open limestone channel method on a laboratory scale. AMD with an initial pH of 2.44 mixed with large-sized limestone particles (11 mm) resulted in pH levels between 2.93 and 3.40. Medium-sized particles (7.12 mm) increased pH to a range of 5.72–6.53, while small particles (4.05 mm) raised the pH to between 6.25 and 6.51.

Furthermore, Hilwani et al. (2022) enhanced the open limestone channel method by incorporating zeolite. Using synthetic AMD with a pH of 2.50, the combination of limestone and zeolite increased the pH to 7.92 at the inlet and 7.20 at the outlet of the channel. Artidarma et al. (2021) employed a slow sand filter with a sand layer thickness of 110 cm to treat river water from the Kapuas River in West Kalimantan, which initially had a pH of 5.60. Using a down-flow filtration system with coastal and silica sand, they successfully raised the water’s pH to between 6.90 and 7.10. In efforts to mitigate the negative impacts of mining activities, it is essential to develop methods that utilize natural materials for AMD management and neutralization before discharge into surface water bodies (Kevinstiv et al., 2023; Luis et al., 2024; Ricardo et al., 2024). Examples of AMD dewatering systems through to final discharge are illustrated in Figures 4 and 5.

Figure 4: Example of an open channel system and water sampling station.

Figure 5: Clean water that has been neutralized from AMD is stored in swamp areas for surface water reserves.

This study builds upon previous research, aiming to utilize materials such as fly ash, zeolite, limestone, and an additional combination with silica sand—an approach that has not been extensively explored. The motivation behind this research is to enhance neutralization efficiency through innovative methods that make use of locally available materials. The study conducted an experiment using neutralizing agents including 100 grams of fly ash, zeolite, and limestone with a channel slope of 5°, successfully increasing AMD pH from 3.75 to a range of 6.00–6.86 (Rizky et al., 2024). Based on these results, further research was carried out using additional neutralizing material – silica sand – as well as different quantities of fly ash and channel slopes, to evaluate the reliability of the open limestone channel method and the effectiveness of various material combinations (Rizky et al., 2024; Pingping et al., 2024; Yu et al., 2024). The passive treatment method, such as the open limestone channel, was selected for its low operational cost, environmental friendliness, and suitability for remote locations. This motivation aligns with the context of mining operations in Indonesia, which often lack the resources required for active treatment systems.

2. MATERIAL AND METHODS

2.1 Research Location

The AMD used in this study was sourced from a sump located near a coal stockpile area within an abandoned mining site. The mining operation was previously managed by a coal mining company situated in the Perambahan area, Salak Village, Talawi Subdistrict, Sawahlunto, West Sumatra, Indonesia, as illustrated in Figure 6.

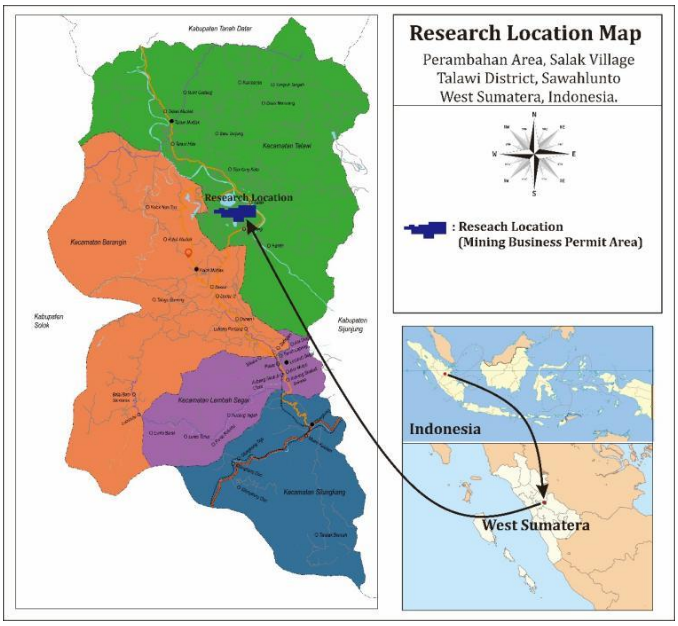

Figure 6: Map of research location

From observational results, the initial pH of the water in the sump at this location was found to average 3.75. This value is below the clean water quality standard threshold established in Indonesia. Therefore, efforts are needed to address this AMD issue to prevent further damage to the region's aquatic ecosystem.

2.2 Testing Method and Experimental Scheme

The Acid Mine Drainage (AMD) testing in this study applied a passive treatment model known as the open limestone channel, designed with eight compartments. This model consists of an open channel constructed with specific dimensions and slopes, as shown in Figure 7. Each compartment is equipped with a baffle to maximize contact time between the neutralizing material and the AMD, while also preventing material migration between compartments. Three different combinations of neutralizing materials were tested, and each combination underwent three separate test runs. pH measurements were taken at designated sampling points in each compartment during the treatment process. The tested combinations included: Combination A (A0, A1, and A2) with a channel slope of 5°, Combination B (B0, B1, and B2) with a 7° slope, and Combination C (C0, C1, and C2) with a 9° slope. The specifications of the neutralizing materials used in each compartment and combination varied, as detailed in Table 1 and illustrated in Figure 7.

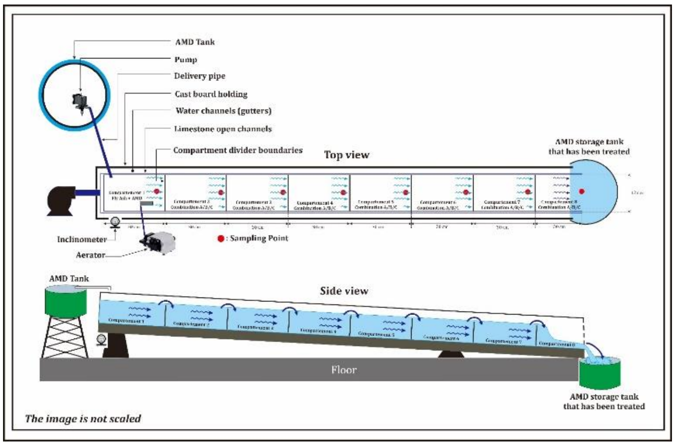

Figure 7: Passive open limestone channel method tool and AMD pH neutralization test scheme.

2.3 Specification of Test Materials

Based on the neutralizing materials used, the differences among the three combinations are as follows: In Combination A, all neutralizing materials were new and had not been previously used in any tests. In Combination B, the materials were reused from Combination A and had been rinsed with acidic water at the test site. Meanwhile, Combination C used materials from Combination B without rinsing. To enhance the effectiveness of AMD neutralization, the materials – limestone, fly ash, zeolite, and silica sand – were first weighed and sieved to obtain an ideal grain size distribution. To remove impurities and reduce moisture content, the materials were washed and then dried under sunlight (Xin et al., 2023; Amira et al., 2022; Sri et al., 2023).

The operation of the open limestone channel system is as follows: In Combination A, AMD from the storage tank was pumped into Compartment 1 containing fly ash. It then flowed into Compartment 2 containing Zeolite A (grain size 16.0–25.0 mm, weight 1,500 grams), followed by Compartment 3 with Zeolite B (8.0–16.0 mm, 1,500 grams). The flow continued into Compartment 4 filled with Silica Sand A (1.18–2.36 mm, 2,200 grams) and Compartment 5 with Silica Sand B (0.6–2.0 mm, 2,200 grams). It then moved to Compartment 6 containing Limestone A (1.18–25.0 mm, 2,400 grams), and continued into Compartment 7 with Limestone (4.0–19.0 mm, 2,400 grams). The final compartment, Compartment 8, contained no neutralizing material. The test results for Combination A are presented in Table 2. The same configuration applies to Combinations B and C, as guided by Table 1, with their respective results shown in Tables 3 and 4. During the testing process, the limestone dissolves in the AMD and contributes alkalinity that functions as a neutralizing agent (Paul et al., 1997; Casey et al., 2022; Kai et al., 2022). This alkalinity interacts with the neutralizing agents (fly ash, zeolite, and silica sand) in each compartment through which the AMD flows.

Table 1: Specification for Acid Mine Drainage (AMD) pH Neutralization Test Materials.

Compartment Number	Neutralizing Agents	Sizes (mm)	Material Weight (gram)	Channel Slope
Combination A				
	Acid Mine Drainage (AMD)	–	–	**5°**

Table 1(Cont.): Specification for Acid Mine Drainage (AMD) pH Neutralization Test Materials.

1	AMD + Fly Ash	–	100.0	
2	Zeolite (A)	16.0 - 25.0	1,500.0	
3	Zeolite (B)	8.0 - 16.0	1,500.0	
4	Silica Sand (A)	1.18 - 2.36	2,200.0	
5	Silica Sand (B)	0.6 - 2.0	2,200.0	
6	Limestone (A)	1.18 - 3.35 and 25.0	2,400.0	
7	Limestone (B)	4.0 - 19.0	2,400.0	
8	No neutralizing agent	–	–	
		Combination B		
	Acid Mine Drainage (AMD)	–	–	**7°**
1	AMD + Fly Ash	–	125.0	
2	Silica Sand (A)	1.18 – 2.36	2,200.0	
3	Silica Sand (B)	0.6 - 2.0	2,200.0	
4	Limestone (A)	1.18 - 3.35 and 25.0	2,400.0	
5	Limestone (B)	4.0 - 19.0	2,400.0	
6	Zeolite (A)	16.0 - 25.0	1,500.0	
7	Zeolite (B)	8.0 - 16.0	1,500.0	
8	No neutralizing agent	–	–	
		Combination C		
	Acid Mine Drainage (AMD)	–	–	9
1	AMD + Fly Ash	–	150.0	
2	Limestone (A)	1.18 - 3.35 and 25.0	2,400.0	
3	Limestone (B)	4.0 - 19.0	2,400.0	
4	Zeolite (A)	16.0 - 25.0	1,500.0	
5	Zeolite (B)	8.0 - 16.0	1,500.0	
6	Silica Sand (A)	1.18 – 2.36	2,000.0	
7	Silica Sand (B)	0.6 - 2.0	2,000.0	
8	No neutralizing agent	–	–	

3. RESULTS

3.1 Test Results for Combination A

Combination A was tested three times (A-0, A-1, and A-2) using a compartment slope of 5°. The mixing of AMD with 100 grams of fly ash in Compartment 2 was aided by an aerator and manual stirring to optimize mixing and minimize fly ash sedimentation (Gitari et al., 2006; Viswanath et al., 2012; Abraham et al., 2024).

a. Combination A-0

The test results for Combination A-0 showed an increase in pH in Compartment 2 (A1), indicating that the addition of fly ash effectively raised the AMD pH from 3.75 to 3.93. As AMD flowed from Compartment 2 (A1) into zeolite-filled Compartments 3 (A2) and 4 (A3), the pH increased further to 4.01 and 4.28, respectively. Subsequently, the AMD came into contact with silica sand in Compartments 5 (A4) and 6 (A5), which elevated the pH to 6.86 and 7.39. The flow continued to limestone-filled Compartments 7 (A6) and 8 (A7), resulting in additional pH increases to 7.48 and 7.57. However, in the final Compartment 9 (A8), which contained no neutralizing material, the pH decreased from 7.57 (A7) to 6.69. The pH measurement results for Combination A-0 are presented in Figure 8.

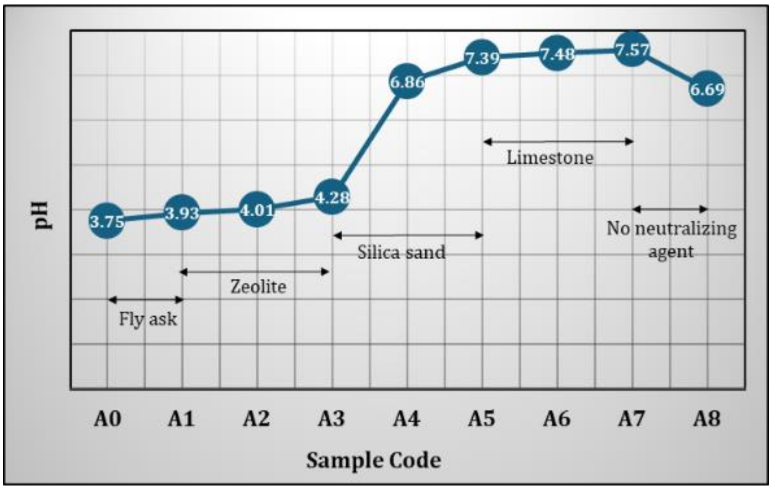

Figure 8: Results of Measuring the pH of Acid Mine Drainage (AMD) in Combination A-0.

b. Combination A-1

The trial for Combination A-1 was conducted with the addition of fly ash. The test results indicated an increase in AMD pH in Compartment 2 (A11), from 3.75 to 4.01. As AMD exited Compartment 2 (A11) and entered zeolite-filled Compartments 3 (A21) and 4 (A31), the pH rose further to 4.91 and 5.08, respectively. The AMD then came into contact with silica sand in Compartments 5 (A41) and 6 (A51), leading to an increase in pH to 7.03 and 7.30. When the AMD flowed into limestone-filled Compartments 7 (A61) and 8 (A71), the pH remained stable at 7.30 in A61 but slightly decreased to 7.21 in A71. In the final Compartment 9 (A81), which did not contain any neutralizing material, the pH dropped significantly to 6.24. The overall pH measurement results for Combination A-1 are shown in Figure 9.

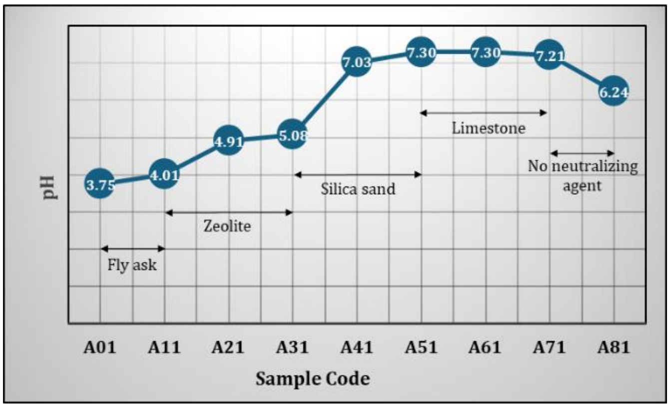

Figure 9: Results of Measuring the pH of Acid Mine Drainage (AMD) in Combination A-1

c. Combination A-2

The test results for Combination A-2, which also used fly ash, are presented in Figure 10. In this combination, the AMD pH in Compartment 2 (A12) increased from 3.75 to 4.01. From there, AMD came into contact with zeolite in Compartments 3 (A22) and 4 (A32), where the pH increased to 4.37 and 4.91, respectively. The AMD then flowed through silica sand in Compartments 5 (A42) and 6 (A52), with corresponding pH increases to 7.30 and 7.66.

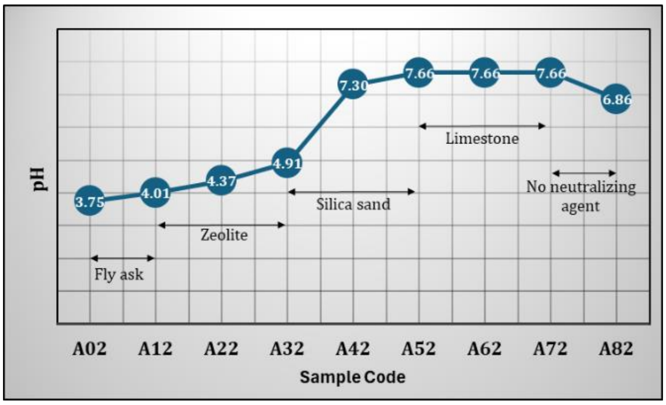

Figure 10: Results of Measuring the pH of Acid Mine Drainage (AMD) in Combination A-2

As AMD exited Compartment 6 (A52) and entered the limestone-filled Compartments 7 (A62) and 8 (A72), the pH remained stable at 7.66 in both compartments. However, in the final Compartment 9 (A82), which did not contain any neutralizing material, the pH dropped to 6.86. A summary of the AMD pH measurements for Combinations A-0, A-1, and A-2 is provided in Table 2.

According to several studies, the decrease in AMD pH observed in the final compartment of this experiment is attributed to the depletion of alkaline or basic materials at the end of the channel (Viswanath et al., 2012; Rusli et al., 2022 & 2023; Abraham et al., 2024). Additionally, AMD coming into contact with oxygen in the final compartment – where no alkaline materials were added—may have led to further oxidation reactions (Gitari et al., 2006; Rusli et al., 2024; Abraham et al., 2024). Another contributing factor is the potential formation of gypsum compounds, which are residual by-products of lime (CaO) reactions that have not fully stabilized (Octiana et al., 2015; Bennetta et al.; Yu et al., 2024; Rusli et al., 2023). These compounds may dissociate, allowing sulfate ions to bind with hydrogen ions present in the water, causing the AMD to become acidic again. For example, in an active treatment study using quicklime over eight weeks, pH values were observed to decrease progressively from 7.35 to 7.22, 6.97, 6.34, and finally to 6.57 (Gitari et al., 2006; Manuel et al., 2011; Banerjee, 2014; Octiana et al., 2015). Therefore, AMD requires sufficient alkalinity as a buffering system to maintain stable pH levels and prevent re-acidification (Asif et al., 2016; Wilson et al., 2018; Youzheng et al., 2020; Pingping et al., 2024).

Table 2: Recapitulation of pH Value Measurement Results Using Combination A.

Compartment Number	Neutralizing Agents	Combination A-0		Combination A-1		Combination A-2		Channel Slope
		Sample Code	pH Measurement Results A-0	Sample Code	pH Measurement Results A-1	Sample Code	pH Measurement Results A-2	
	Acid Mine Drainage (AMD)	A0	3.75	A01	3.75	A02	3.75	5°
1	AMD + Fly Ash	A1	3.93	A11	4.01	A12	4.01	
2	Zeolite (A)	A2	4.01	A21	4.91	A22	4.37	
3	Zeolite (B)	A3	4.28	A31	5.08	A32	4.91	
4	Silica Sand (A)	A4	6.86	A41	7.03	A42	7.30	
5	Silica Sand (B)	A5	7.39	A51	7.30	A52	7.66	
6	Limestone (A)	A6	7.48	A61	7.30	A62	7.66	
7	Limestone (B)	A7	7.57	A71	7.21	A72	7.66	
8	The final result. No neutralizing agent.	A8	6.69	A81	6.24	A82	6.86	

Calcite ($CaCO_3$), a mineral with a positive molar transfer value, can dissolve in acidic water, releasing carbon dioxide (CO_2) gas in the process (Firman et al., 2021; Akhyar et al., 2023; Lei et al., 2024). The concentration of dissolved CO_2 becomes a critical factor influencing pH reduction, as CO_2 increases hydrogen ion concentrations, thus lowering AMD pH (Evgenia et al., 2015; Asif et al., 2021; Firman et al., 2021). High levels of dissolved CO_2 naturally lead to acidic conditions (Asif et al., 2021; Xin et al., 2023). Interestingly, dissolved CO_2 can also facilitate the dissolution of limestone by promoting the reaction between $CaCO_3$ and CO_2 to form bicarbonate products (Barnaby et al., 2005; Evgenia et al., 2015; Asif et al., 2021).

Continuous flow and CO_2 retention can enhance the dissolution or release of limestone, thereby increasing alkalinity production, as the dissolution rate tends to rise with increasing partial pressure of CO_2 (PCO_2) and decreasing pH (Cravotta III et al., 2008; Omar et al., 2014). The aeration process can remove dissolved CO_2 (CO_2 outgassing) from AMD, thereby promoting the conversion of ferrous iron (Fe^{2+}) to ferric iron (Fe^{3+}) (Evgenia et al., 2015; Gurkiran et al., 2018; Kleinmann & Ackman, 2024). In this study, the selection of materials in Combination A was based on previous research, which demonstrated the potential of fly ash, silica sand, zeolite, and limestone to increase water pH, particularly when applied in an open limestone channel system for AMD treatment (Hilwani et al., 2022; Wang et al., 2024; Hu et al., 2024; Goumih et al., 2025).

3.2 Test Results for Combination B

Combination B was tested three times (B-0, B-1, and B-2) with a compartment slope of 7°. In each test, AMD was mixed with 100 grams of fly ash in Compartment 2, using an aerator and manual stirring to optimize mixing and minimize fly ash sedimentation (Gitari et al., 2006; Viswanath et al., 2012; Abraham et al., 2024).

a. Combination B-0

The results of the AMD test using Combination B-0 showed a pH increase in Compartment 2 (B1). The addition of fly ash proved effective in raising the AMD pH from 3.75 to 4.01. As AMD flowed into zeolite-filled Compartments 3 (B2) and 4 (B3), the pH continued to rise to 4.91 and 5.17, respectively. Further contact with silica sand in Compartments 5 (B4) and 6 (B5) resulted in higher pH values of 6.86 and 7.39. However, when the AMD entered the limestone-filled Compartments 7 (B6) and 8 (B7), the pH dropped to 5.53 and 5.62. In the final Compartment 9 (B8), which contained no neutralizing material, the pH decreased further, reaching 5.53. The pH measurement results for Combination B-0 are presented in Figure 11.

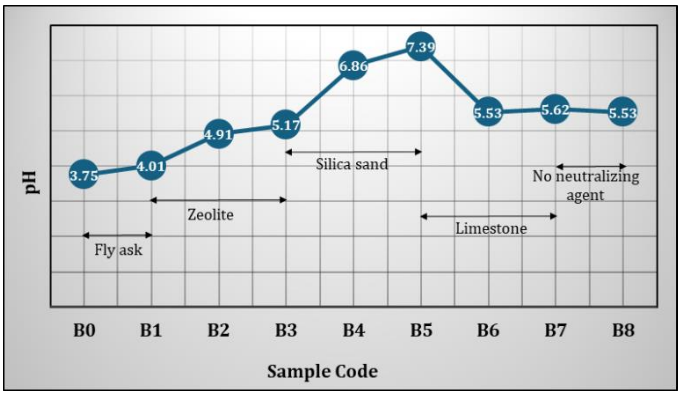

Figure 11: Results of Measuring the pH of Acid Mine Drainage (AMD) in Combination B-0

b. Combination B-1

In the trial of Combination B-1, an increase in AMD pH was observed in Compartment 2 (B11). The addition of fly ash in this compartment successfully raised the pH from 3.75 to 4.01. After passing through Compartment 2 (B11), the AMD came into contact with silica sand in Compartments 3 (B21) and 4 (B31). The results showed that the pH increased significantly to 7.12 and 7.30, respectively. Following this, AMD flowed through limestone in Compartments 5 (B41) and 6 (B51), where the pH remained stable at 7.30. The next contact occurred in zeolite-filled Compartments 7 (B61) and 8 (B71), which caused the pH to decrease to 5.97 and 5.71. In the final Compartment 9 (B81), which contained no neutralizing agent, the AMD pH slightly increased to 5.89. The overall pH measurement results for Combination B-1 are presented in Figure 12.

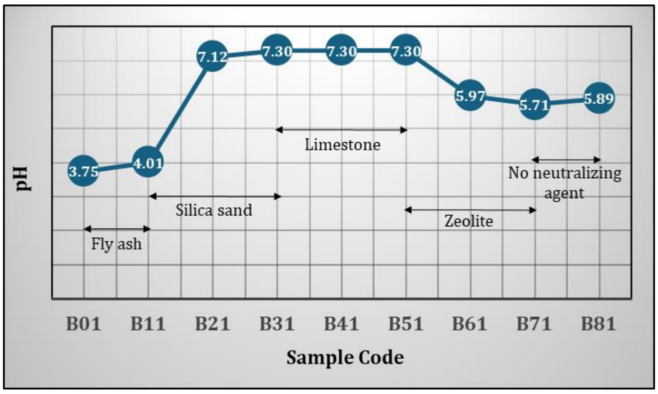

Figure 12: Results of Measuring the pH of Acid Mine Drainage (AMD) in Combination B-1

c. Combination B-2

In the trial of Combination B-2 using fly ash, the AMD pH in Compartment 2 (B12) increased from its initial value of 3.75 to 4.01. The complete pH measurement results for this combination are presented in Figure 13. After leaving Compartment 2 (B12), AMD came into contact with silica sand in Compartments 3 (B22) and 4 (B32), where the pH increased to 7.12 and 7.39, respectively. Subsequently, AMD interacted with limestone in Compartments 5 (B42) and 6 (B52).

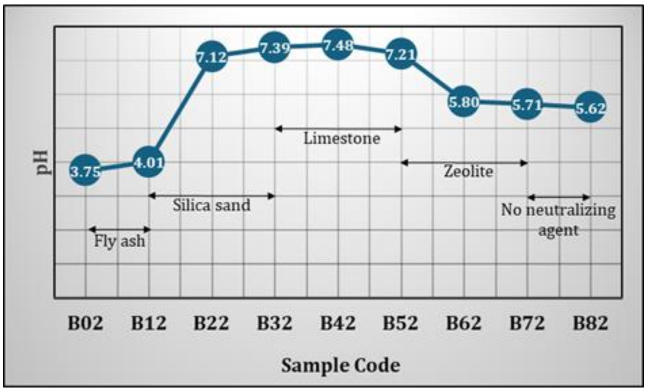

Figure 13: Results of Measuring the pH of Acid Mine Drainage (AMD) in Combination B-2

In Compartment 5 (B42), the AMD pH increased to 7.48. However, in Compartment 6 (B52), the pH dropped to 7.21. The AMD exiting Compartment 6 (B52) then came into contact with zeolite in Compartments 7 (B62) and 8 (B72), where the pH further decreased to 5.80 and 5.71, respectively. In the final Compartment 9 (B82), which did not contain any neutralizing material, a slight pH decrease was observed, reaching 5.62. A summary of the pH measurements for Combinations B-0, B-1, and B-2 is presented in Table 3.

Table 3: Recapitulation of pH Value Measurement Results Using Combination B.

Compartment Number	Neutralizing Agents	Combination B-0		Combination B-1		Combination B-2		Channel Slope
		Sample Code	pH Measurement Results B-0	Sample Code	pH Measurement Results B-1	Sample Code	pH Measurement Results B-2	
	Acid Mine Drainage (AMD)	B0	3.75	B01	3.75	B02	3.75	**7°**
1	AMD + Fly Ash	B1	4.01	B11	4.01	B12	4.01	

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Table 3(Cont.): Recapitulation of pH Value Measurement Results Using Combination B.

2	Silica Sand (A)	B2	4.91	B21	7.12	B22	7.12	
3	Silica Sand (B)	B3	5.17	B31	7.30	B32	7.39	
4	Limestone (A)	B4	6.86	B41	7.30	B42	7.48	
5	Limestone (B)	B5	7.39	B51	7.30	B52	7.21	
6	Zeolite (A)	B6	5.53	B61	5.97	B62	5.80	
7	Zeolite (B)	B7	5.62	B71	5.71	B72	5.71	
8	The final result. No neutralizing agent.	B8	5.53	B81	5.89	B82	5.62	

The observed decrease in AMD pH during the Combination B tests can be attributed to variations in experimental treatment. The alkaline fly ash material did not fully mix with the AMD due to sedimentation and filtration by subsequent neutralizing materials, resulting in pH reductions in downstream compartments (Sari et al., 2020; Ricardo et al., 2024). Additionally, the pH decline observed during the zeolite phase may be due to the ineffective activity of sulfate-reducing bacteria, which may have been suppressed by impurities in the zeolite material (Gitari et al., 2007; Sari et al., 2020; Viswanath et al., 2012).

3.3 Test Results for Combination C

Combination C was tested three times (C-0, C-1, and C-2) using a compartment slope of 9°. In each test, AMD was mixed with 100 grams of fly ash in Compartment 2, aided by an aerator and manual stirring to enhance mixing and reduce fly ash sedimentation (Gitari et al., 2006; Viswanath et al., 2012; Abraham et al., 2024).

a. Combination C-0

In the C-0 trial using fly ash, an increase in AMD pH was observed in Compartment 2 (C1), rising from 3.75 to 4.19. From Compartment 2 (C1), AMD flowed into limestone-filled Compartments 3 (C2) and 4 (C3), where the pH increased to 5.80 and 6.15, respectively. From Compartment 4 (C3), AMD entered zeolite-filled Compartments 5 (C4) and 6 (C5), resulting in further pH increases to 6.42 and 6.86. Subsequently, AMD exiting Compartment 6 (C5) flowed into silica sand-filled Compartments 7 (C6) and 8 (C7). The pH in Compartment 7 (C6) rose to 7.39, but dropped again in Compartment 8 (C7) to 6.86. In the final Compartment 9 (C8), which did not contain any neutralizing material, the pH further decreased to 6.51. The pH measurement results for Combination C-0 are shown in Figure 14.

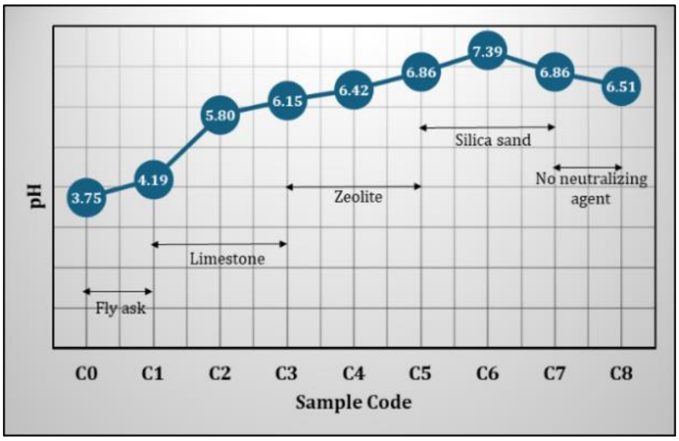

Figure 14: Results of Measuring the pH of Acid Mine Drainage (AMD) in Combination C-0.

b. Combination C-1

In the Combination C-1 trial, the addition of fly ash resulted in an increase in AMD pH from 3.75 to 4.28 in Compartment 2 (C11). Subsequently, AMD from Compartment 2 (C11) came into contact with limestone in Compartments 3 (C21) and 4 (C31). In these two compartments, the pH increased to 6.42 in Compartment 3 (C21) but then slightly decreased to 6.24 in Compartment 4 (C31). From Compartment 4 (C31), AMD continued to interact with zeolite in Compartments 5 (C41) and 6 (C51). The pH decreased to 5.26 in Compartment 5 (C41), but increased again to 5.62 in Compartment 6 (C51). The next treatment involved silica sand in Compartments 7 (C61) and 8 (C71), where the pH in both compartments increased to 6.86. In the final Compartment 9 (C81), which did not contain any neutralizing material, the AMD pH dropped slightly to 6.33. The pH measurement results for Combination C-1 are presented in Figure 15.

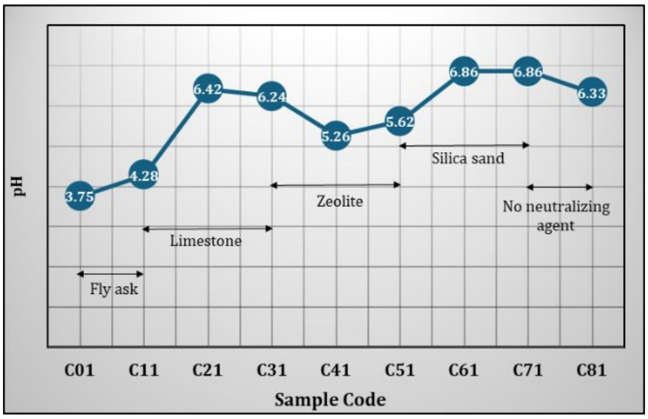

Figure 15: Results of Measuring the pH of Acid Mine Drainage (AMD) in Combination C-1.

c. Combination C-2

In the Combination C-2 test using fly ash (Figure 16), the AMD pH in Compartment 2 (C12) increased from 3.75 to 4.28. With the addition of limestone in Compartments 3 (C22) and 4 (C32), the pH further rose to 5.71 and 6.06, respectively. From Compartment 4 (C32), AMD contacted zeolite in Compartments 5 (C42) and 6 (C52), where the pH increased to 6.15 in C42 but then decreased to 5.71 in C52. Next, AMD passed through silica sand in Compartments 7 (C62) and 8 (C72), where the pH increased to 7.03 and 7.21, respectively. In the final Compartment 9 (C82), which lacked neutralizing material, the pH decreased again to 6.60.

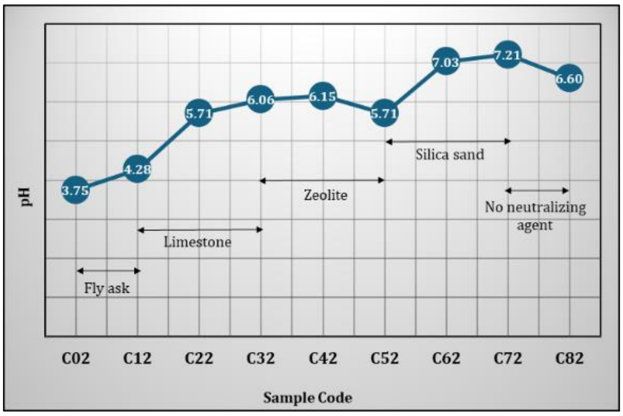

Figure 16: Results of Measuring the pH of Acid Mine Drainage (AMD) in Combination C-2.

A summary of pH measurements for Combinations C-0, C-1, and C-2 is presented in Table 4. The highest pH achieved in Combination C was 7.21, observed in Compartment 7. However, in Compartment 9, the pH decreased again to 6.60. This suggests that AMD can be discharged into the surrounding aquatic environment only if the pH remains above the permissible limit.

Table 4: Recapitulation of pH Value Measurement Results Using Combination C

Compartment Number	Neutralizing Agents	Combination C-0		Combination C-1		Combination C-2		Channel Slope
		Sample Code	pH Measurement Results C-0	Sample Code	pH Measurement Results C-1	Sample Code	pH Measurement Results C-2	
	Acid Mine Drainage (AMD)	C0	3.75	C01	3.75	C02	3.75	9°
1	AMD + Fly Ash	C1	4.19	C11	4.28	C12	4.28	
2	Limestone (A)	C2	5.80	C21	6.42	C22	5.71	
3	Limestone (B)	C3	6.15	C31	6.24	C32	6.06	
4	Zeolite (A)	C4	6.42	C41	5.26	C42	6.15	
5	Zeolite (B)	C5	6.86	C51	5.62	C52	5.71	
6	Silica Sand (A)	C6	7.39	C61	6.86	C62	7.03	
7	Silica Sand (B)	C7	6.86	C71	6.86	C72	7.21	
8	The final result. No neutralizing agent.	C8	6.51	C81	6.33	C82	6.60	

The pH reduction observed during the Combination C tests may be attributed to several factors, including the depletion of alkaline material in the final compartments, the formation of surface coatings on the limestone due to impurities, and reduced absorption and reactivity on the limestone surface (Kumar et al., 2008; Hao et al., 2021; Acharya & Kharel, 2020). The particle size of the limestone also significantly affects its neutralization efficiency; larger particles (20–40 mm) are generally less effective than smaller ones (5–10 mm) in neutralizing AMD (Alcolea et al., 2012; Ioan et al., 2017; Hugh et al., 2011).

The test results also indicated that contact time between AMD and limestone is a critical factor. Adjusting the channel length, slope, and flow velocity can increase the contact time, improving neutralization efficiency. Retention efficiencies for Cd and Fe were especially notable during the first hour of testing, and similar trends were observed in the limestone's capacity to neutralize SO_4^{2-} (Kumar et al., 2008; Alcolea et al., 2012; Acharya & Kharel, 2020; Hilwani et al., 2022; Goumih et al., 2025). In this study, the selection of test materials for Combination C was based on prior research, particularly the use of fly ash, silica sand, zeolite, and limestone in configurations outlined in Table 4. These materials were proven effective for pH enhancement when applied in an open limestone channel system for AMD treatment (Fadhilah et al., 2021; Hilwani et al., 2022; Hu et al., 2024; Goumih et al., 2025).

3.4 Efficiency of pH Increase in AMD Based on Test Results

Efficiency, in the context of pH enhancement in AMD, refers to the optimization of both measurement and treatment processes to achieve accurate, rapid, cost-effective, and low-waste outcomes (Gitari et al., 2006; Manuel et al., 2011). The relevant components include: (1) Measurement efficiency, which covers the accuracy and precision of measuring instruments, as well as speed and real-time responsiveness. (2) Neutralizing material efficiency, which involves dose optimization based on measurement data, and minimization of costs and secondary waste. (3) Treatment process efficiency, which includes the integration of measurement and treatment systems, along with continuous performance evaluation. (4) Environmental and economic efficiency, which entails reducing environmental impacts while balancing costs and benefits (Gitari et al., 2006; Manuel et al., 2011; Banerjee, 2014; Ricardo, 2024). The efficiency results of AMD pH measurements in this study are presented in Table 5.

Table 5: Increase in pH of AMD in Each Experimental Combination

Combination		Initial pH of AMD	Final pH of AMD test results	Efficiency of pH increase	
				Efficiency of each test (%)	Average increase efficiency (%)
A	A-0	3.75	6.69	78	76
	A-1		6.24	66	
	A-2		6.86	83	
B	B-0	3.75	5.53	47	62
	B-1		5.89	57	
	B-2		5.62	50	
C	C-0	3.75	6.51	74	74
	C-1		6.33	69	
	C-2		6.60	76	

If the four factors mentioned above are not taken into account, measurement efficiency cannot be achieved, and the release of AMD into surrounding water bodies may harm aquatic ecosystems. Efficiency in AMD measurement and treatment is a combination of instrument precision, response speed, resource optimization, and environmental sustainability. Without efficiency, efforts to increase pH may become ineffective, wasteful, or even exacerbate existing problems (López et al., 2010; Viswanath et al., 2012). In this study, the efficiency of AMD pH improvement was found to be substantial. The pH increase efficiency measured for Combination A was 76%, for Combination B was 62%, and for Combination C was 73%.

4. DISCUSSION

The effectiveness of material combinations in neutralizing AMD pH in this study demonstrates that locally available materials (fly ash, zeolite, limestone, and silica sand) used in an open limestone channel system can effectively neutralize Acid Mine Drainage (AMD) with an initial pH of 3.75. Combination A, which used entirely new materials, achieved the highest chemical efficiency (76%) with an average final pH of 7.51. However, optimal neutralization occurred in the final compartment (Compartment 8), indicating the need for longer contact time to reach maximum efficiency. This is consistent with findings by Rusli & Samosir (2021), who emphasized the role of fly ash in increasing pH through alkalinity reactions. The drawback of Combination A, however, lies in its reliance on new materials, which may raise operational costs.

Combination B, which reused materials from Combination A, achieved an average pH of 7.39 (62% efficiency), with faster neutralization occurring in Compartments 5–6. This improved rate was due to surface activation of the materials from prior exposure to AMD, enhancing their chemical reactivity. These findings align with Hilwani et al. (2022), who reported enhanced performance of reused zeolite and limestone. Although its chemical efficiency was lower, Combination B was considered technically optimal due to reduced material usage and shorter processing time—critical advantages for AMD treatment in remote areas.

Combination C, which used unrewashed materials from Combination B, showed a final compartment pH drop to 6.60, despite achieving 73% chemical efficiency. This drop was attributed to CO_2 accumulation from calcium carbonate ($CaCO_3$) reactions with acid, and the formation of gypsum residues ($CaSO_4{\cdot}2H_2O$) that clogged material pores. Similar effects were documented by Evgenia et al. (2015), confirming that CO_2 accumulation can lower pH through carbonic acid (H_2CO_3) formation.

Key success factors and challenges observed in this study include: (1) Particle size and contact time: The limestone grain size significantly influenced neutralization rate. Smaller particles (4–16 mm) in Combination B provided a larger surface area and faster neutralization compared to larger particles (16–25 mm) in Combination A. This supports findings by Nurfasiha & Kusuma (2020), who reported up to 50% higher efficiency with smaller limestone sizes. (2) Microbial activity: The pH decline in final compartments of Combinations B and C is likely due to the activity of Acidithiobacillus ferrooxidans, which oxidizes Fe^{2+} to Fe^{3+} and accelerates pyrite oxidation, thus generating secondary acidity (Stumm & Morgan, 1996). (3) Material pore clogging: The formation of gypsum and iron hydroxide coatings on limestone surfaces reduced material reactivity, a major limitation in passive systems, as also reported by Cravotta III et al. (2008) and Abrahams et al. (2024).

Practical Implications and Recommendations: This study emphasizes the need to integrate chemical and technical aspects in AMD treatment system design. While Combination A achieved the highest pH, its use of new materials and longer processing time are less feasible for large-scale applications. In contrast, Combination B offers a more sustainable solution by reusing materials, reducing secondary waste, and optimizing costs. To enhance performance, the following actions are recommended: (1) Material Pretreatment: Washing zeolite and limestone before use to remove impurities that hinder reactions. (2) Aeration System: Installing aeration in the final compartments to reduce CO_2 accumulation and oxidize Fe^{2+} prior to discharge. (3) Routine Monitoring: Regular pH monitoring and replacement of clogged materials to maintain efficiency.

Limitations in this study did not assess the long-term performance of the materials under continuous flow conditions, particularly regarding heavy metal accumulation and pH stability. Additionally, the use of synthetic AMD with specific characteristics may not represent the variability of real AMD in different locations. Therefore, Combination B emerges as the optimal strategy for AMD neutralization, balancing chemical performance (pH 7.39), processing speed, and material efficiency. This approach is especially relevant to mining operations in Indonesia that prioritize low-cost, environmentally friendly solutions. Future studies are encouraged to explore integration with active treatment methods (e.g., electrocoagulation) to enhance system sustainability.

CONCLUSION

This study evaluated the effectiveness of three treatment combinations (A, B, and C) in neutralizing the pH of Acid Mine Drainage (AMD), which initially had a pH of 3.75. The results revealed distinct pH elevation patterns and neutralization efficiencies among the combinations, each influenced by specific factors. In Combination A, the final pH values were: A-0 = 7.57; A-1 = 7.30; A-2 = 7.66, with an average of 7.51 – an increase of 3.76 units from the initial value. The neutralization efficiency was 76%, and the mechanism occurred when AMD reached Compartment 8, which contained limestone. In Combination B, final pH values were: B-0 = 7.39; B-1 = 7.30; B-2 = 7.48, with an average of 7.39 – an increase of 3.64 units. The efficiency was 62%, with neutralization occurring earlier in Compartments 5 and 6, also filled with limestone. For Combination C, final pH values were: C-0 = 7.39; C-1 = 6.86; C-2 = 7.21, with an average of 7.15 – an increase of 3.40 units. The neutralization efficiency was 73%, and the process took place in Compartments 7 and 8. Despite Combinations A and C demonstrating higher chemical efficiency (76% and 73%, respectively), Combination B was determined to be the most practically effective. This is due to its faster neutralization process, which began earlier in the channel, and more efficient use of limestone material without compromising the pH increase (average 7.39). Factors contributing to result variations included: Gypsum precipitate formation reducing limestone reactivity; CO_2 gas accumulation hindering neutralization reactions, and Pore clogging of test materials by residues and surface coatings that limited AMD contact. The efficiency ranking by parameter was as follows: Chemical efficiency (pH increase): A (76%) > C (73%) > B (62%). Technical efficiency (time and material use): B > A > C. This study confirms that the effectiveness of AMD neutralization depends not only on pH increase but also on operational efficiency. Combination B emerges as the optimal choice due to its balance between sufficient pH elevation (7.39) and more efficient resource utilization. These findings recommend the integration of technical and chemical considerations in the design of AMD treatment systems.

AUTHOR CONTRIBUTIONS

Rusli HAR: Conceptualization, Research methodology, Data curation, Data analysis, Supervision, Project administration; Ichsan Invanni Baharuddin & Riam Marlina A: Writing research drafts, editing, finalizing articles; Adree Octova & Aulia Hidayat B: Grammar and scientific writing techniques; Rangga Agung P.H., Ahmad Fadly & Rudi Anarta: Data analysis, quality control, writing the research draft; Riko Maiyudi, Harizona A. R., & Rizky Ananda: Surveyor, collecting research data, designing research equipment, testing.

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