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The Effect of the Comparison of Zeolite and Limestone on the Degree of Acidity (pH) of Acid Mine Drainage (Case Study: PT. XYZ)

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Abstract: Acid Mine Drainage (AMD) is a serious environmental problem in the particularly Minerals mining industry, characterized by low pH and high concentrations of dissolved heavy metals. This study aims to analyze the effect of the mass ratio between natural zeolite and limestone as a neutralization medium and adsorbent in the AMD processing of PT. XYZ. The research method used was a batch experiment in the laboratory with five variations of the mass ratio of zeolite and limestone (100:0, 75:25, 50:50, 25:75, and 0:100) with a total mass of 10 grams per 500 mL of AMD sample. The parameter measured was the change in pH during a contact time of 90 minutes. The results showed that all variations were able to increase the AMD pH from an initial value of 2.6. The ratio of 25:75 (zeolite:limestone) showed the most optimal effectiveness, able to increase the pH to 8.02 in 60 minutes and remained stable to time 90 minutes. Limestone plays a dominant role in acid neutralization, while the presence of zeolite is thought to prevent the effects of zeolite on acid neutralization armouring (coating) on limestone particles and also absorbs existing metal ions. This combination proved more effective than using each ingredient alone. It was concluded that a mass ratio of 25:75 was the optimal ratio for AMD treatment in this case study.

Kata Kunci: Acid Mine Drainage (AMD), Zeolite, Limestone, Process Batch

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Introduction

The Republic of Indonesia has long been known for its abundance of natural resources, including in the fields of plantations, agriculture, fisheries, and mining. This attraction is also the cause of the arrival of colonial countries to the archipelago. Almost all parts of the Republic of Indonesia have natural resources that can provide benefits for the well-being of the people. One of the natural resources that can improve the standard of living of the community is the field of mining, both traditional and industrial (Nurfatimah, 2023).

The mining industry, particularly Minerals, has a vital role in the national economy, but it also generates a significant environmental impact. Although it provides good benefits, this mining activity also has a negative impact (Tanri et al., 2024). One of the main impacts is the formation of Acid Mine Water or AMD, which is water that has a high level of acidity, which is produced from the mining process and rich in dissolved metals (Ananda et al., 2024). AMD is formed when sulfide minerals, such as pyrite (FeS_2), which are exposed to air and water during the mining process, undergo oxidation (Zipper et al., 2018). This reaction produces sulfuric acid that drastically lowers the pH of the water, often below 4, and dissolves heavy metals such as iron (Fe), manganese (Mn), and aluminum (Al) from surrounding rocks (Sun et al., 2018).

The release of AMD into water bodies without adequate treatment can cause damage to aquatic ecosystems, kill aquatic biota, and pollute water sources for humans (Johnson & Hallberg, 2005). Therefore, the government sets strict environmental quality standards for mine wastewater, where the pH parameter must be in the range of 6-9 before it can be discharged into the environment (Minister of Environment Decree 2003). One of the negative consequences of mining activities is the emergence of Acid Mine Drainage (AMD), which is water

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with a high acidity level, produced by the mining process and rich in dissolved metals. Similarly, acid mine drainage (AMD) (Ananda et al., 2024).

Managing acidic water produced by mining, whether in active or passive forms, is crucial to achieving environmental quality standards. This process involves adjusting the pH value, as well as the levels of Fe and Mn, as stipulated in Minister of Environment Regulation No. 113 of 2003 concerning wastewater quality standards from mining activities, with a pH value set between 6 and 9 (Minister of Environment Decree, 2003).

Various technologies have been developed to treat AMD, which are generally divided into two categories: active and passive treatment (Mapukata et al., 2024). Active treatment, such as the addition of alkaline chemicals (quicklime, caustic soda), is effective but requires high operational and energy costs, as well as continuous monitoring (Thisani et al., 2021). In contrast, passive treatment systems are an attractive alternative due to their environmental friendliness, low cost, and minimal maintenance (Qinwen et al., 2024).

One potential passive treatment method is the use of naturally occurring reactive materials. Limestone (predominantly $CaCO_3$) has long been used for its effective acidity neutralization (Francesco et al., 2015). However, the use of limestone alone has a drawback, namely the phenomenon of armouring, where the surface of limestone particles is covered by metal hydroxide deposits (e.g. $Fe(OH)_3$), thus inhibiting further neutralization reactions (Sun et al., 2018).

To address these issues, modern research has begun combining limestone with other materials, one of which is zeolite. Zeolite is a hydrated aluminosilicate mineral with a porous three-dimensional framework structure. Its unique structure provides a high cation exchange capacity, enabling it to adsorb heavy metal ions from AMD solutions (Widyaningrum et al., 2022). Furthermore, the porosity of zeolite is expected to act as a dispersion medium for limestone, reducing direct contact between limestone particles and minimizing the armoring effect (Sun et al., 2018).

Although the potential of each material has been extensively researched, studies on the effect of the compositional ratio between zeolite and limestone on the effectiveness of AAT neutralization are still limited. Therefore, this study aims to determine the optimal mass ratio between zeolite and limestone to increase the pH of acid mine water originating from the PT. XYZ site, thus providing the basis for the development of a more efficient and sustainable passive treatment system (Daniela et al., 2025).

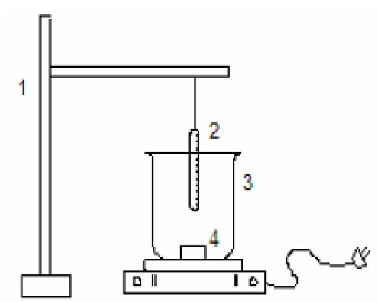
Method

Time and Place of Research

This research was conducted at the Chemical Engineering Laboratory, Bontang Industrial Technology College from March – May 2025. Acid Mine Drainage samples were obtained from the reservoir pond (settling pond) in the concession area of PT. XYZ, South Sulawesi.

Tools and Materials

The equipment used includes a digital pH meter (Hanna Instruments HI98191), 1000 mL beaker, analytical balance, magnetic stirrer, Whatman No. 42 filter paper, thermometer and stopwatch. The materials used are samples of AMD PT. XYZ, natural zeolite type clinoptilolite which has been activated by grinding to a size of 100 mesh and heating at a temperature of 110°C for 2 hours, as well as commercial limestone which is also ground to a size of 100 mesh. The main tools of this study can be seen in Figure 1 below:



Information:
1. Tripod
2. pH meter (Hanna Instruments HI98191),
3. Beaker Glass
4. Magnetic Stirrer

Figure 1. Research Tools Series

Research Methods

Initial characterization of the AMD sample was carried out by measuring the initial pH using a calibrated pH meter. The experimental procedure was carried out in a batch as follows. The first stage is five beakers were prepared, each filled with 500 mL of AMD sample. The next stage is A mixture of zeolite and limestone media was added to each beaker with a total mass of 10 grams, according to five variations in mass ratio (in grams): (V1: 10 g Zeolit + 0 g Limestone (100:0), V2: 7,5 g Zeolit + 2,5 g Limestone (75:25), V3: 5.0 g Zeolite + 5.0 g Limestone (50:50), V4: 2.5 g Zeolite + 7.5 g Limestone (25:75), and the last V5: 0 g Zeolit + 10 g Limestone (0:100). Each solution was stirred using a magnetic stirrer at a constant speed (150 rpm) to ensure even contact between the media and AMD. After that we changes in pH were measured and recorded at time intervals of 0, 5, 15, 30, 60, and 90 minutes. Each treatment was repeated three times (triplicate) to ensure data validity.

The pH measurement data from the three replications were averaged. The results are then presented in the form of tables and graphs showing the relationship between contact time and pH change for each variation of the media comparison. The analysis was carried out descriptively to compare the effectiveness of each variation and determine the most optimal comparison.

Results and Discussion

The results of AMD sample measurements from PT. XYZ showed an average initial pH of 2.6. This value is well below the established environmental quality standards (pH 6-9), requiring treatment before discharge into the environment. Changes in AAT pH during 90 minutes of contact time for the five variations of zeolite and limestone ratios are presented in Table 1.

Table 1. Results of Acid Mine Drainage Measurements After Adding a Mixture of Zeolite and Limestone

Variables	**pH Per Time (Minutes)**					
	0	**5**	**15**	**30**	**60**	**90**
V1	2,6	2,68	3,5	3,65	3,72	3,81
V2	2,6	2,7	3,53	3,88	4,2	5,6
V3	2,6	2,72	3,58	5	7,2	7,26
V4	2,6	2,89	4,3	6,8	8,02	8,02
V5	2,6	3,02	4,5	7,3	7,6	7,6

Source: Karim (2025)

Data Based on Table 1, it can be seen that all treatment variations successfully increased the AMD pH. The most significant pH increase occurred in the first 30 minutes for all variations containing limestone (V2, V3, V4, and V5). This indicates that the neutralization reaction by limestone ($CaCO_3$) occurs relatively quickly. The data from above, if displayed in a graph, will more clearly show the changes in the increase in pH of acid mine water at each time in all variables, as shown in Figure 2 below.

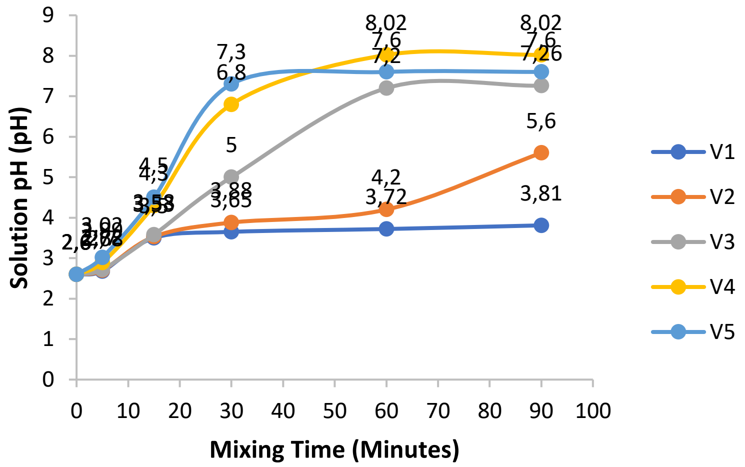


Figure 2. Graph of pH Changes for Each Time Unit Variable

In treatment V1 (100% zeolite), the pH increase was very slow and insignificant, reaching only pH 3.81 after 90 minutes. This confirms that the primary role of zeolite is not as an acid-neutralizing agent, but rather through a cation exchange mechanism where zeolite can absorb limited amounts of H^+ or acid-inducing metal ions such as Al^{3+} and Fe^{3+} (Widyaningrum et al. 2022).

Treatment V5 (100% limestone) demonstrated very rapid neutralization, reaching a pH of 7.3 in 30 minutes. However, after that, the pH increase slowed and tended to stagnate around 7.6. This slowdown is strongly suspected to be due to the initial formation of a passive layer (armouring) from metal hydroxide deposits on the surface of limestone particles, which prevents further contact with AAT (Sun et al., 2018).

Interestingly, the mixture variations showed superior results. Treatment V4 (25% zeolite: 75% limestone) performed best. The pH increased rapidly, reaching 8.02 within 60 minutes, and remained constant at the same value until the end of the observation period at 90 minutes. The high limestone content (75%) provided a large neutralization capacity, while the presence of zeolite (25%) is thought to act as a dispersant. The zeolite particles dispersed among the limestone particles create space and prevent aggregation of the limestone particles, thereby reducing the possibility of armouring massively. Thus, the active surface of the limestone can continue to react with the acid until it reaches a neutral pH.

Treatment V3 (50:50) also showed good results, with a final pH of 7.26, still below V4 but already showing a pH condition that was trending towards good. Meanwhile, V2 (75:25), which has less limestone content, was only able to achieve a final pH of 5.6, still classified as weakly acidic. This indicates that the amount of limestone remains the dominant factor in determining the total neutralization capacity of the system.

The effectiveness of this combination is in line with research by Daniela et al., (2025) which found that zeolite and limestone-based composite materials were able to increase the efficiency of cadmium Cd absorption. In the synergistic treatment of wastewater. Zeolite not only functions as a metal adsorbent but also as a "framework" that maintains the reactivity of the neutralizing material so that a passive layer does not form (armouring) from metal hydroxide deposits on the surface of limestone particles (Sun et al., 2018).

Based on the test results in this study, it can be seen that adding a mixture of zeolite and lime can increase the pH of acidic water produced from AMD. The most significant pH increase occurred in the first 30 minutes for all variations containing limestone (V2, V3, V4, and V5). From the graph, it can be seen that limestone ($CaCO_3$) plays a role in neutralizing acid mine drainage by increasing the pH of the artificial acid water from pH 2.6 to 8.02 when 10 grams of 25% zeolite: 75% limestone. This increase in pH indicates that lime ($CaCO_3$) functions as a neutralizer for acidic water. Next, we will discuss the neutralization reaction that occurs between sulfuric acid and calcium carbonate.

$$2CaCO_3 + 2H_2SO_4 \rightarrow (CaSO_4)_2 + 2H_2O + 2CO_2$$

Here, anhydrous calcium sulfate $(CaSO_4)2$is a salt produced from a strong acid, namely sulfuric acid (H_2SO_4), and a strong base, namely Calcium hydroxide ($Ca(OH)_2$). Calcium hydroxide ($Ca(OH)_2$) is formed through the reaction between Calcium Carbonate ($CaCO_3$) and Water (H_2O), which is often referred to as the hydrolysis reaction of lime (CaCO3). The reaction process is:

$$CaCO_3 + 2H_2O \rightarrow Ca(OH)_2 + H_2CO_3$$

From this reaction, it can be seen that $CO_3^{2-}$from lime ($CaCO_3$) when reacting with H^+ in acidic solution will produce H_2CO_3 and OH^-. The longer the CO_3^{2-} from lime ($CaCO_3$) in contact with H^+ in acidic solution, the number of H+ ions in the solution will decrease, so that the pH of the water becomes higher (acidity decreases) (Metboki, et al., 2018). When lime interacts with acidic water, a hydrolysis reaction occurs, breaking down calcium carbonate into calcium ions (Ca^{2+}) and hydroxide ions (OH^-). In this process, hydroxide ions (OH^-) formed from the hydrolysis of lime increases the OH content-in solution, which further increases the pH of the solution. Furthermore, calcium ions (Ca^{2+}) produced from calcium carbonate interacting with sulfate ions (SO_4^{2-}) which comes from sulfuric acid, then forms calcium sulfate salt which is insoluble in water. (Kusdarini, et al, 2024).

This study is in accordance with previous research conducted by Sari, et al., (2020) found that the use of lime with a dose of 0.2 g/L can increase the change in the initial pH value from 3.43 to 7.11 (Sari et al. 2020). Furthermore, research conducted by Ignasius Nartoris can be found that the use of lime of 0.2 g/L can increase the change in pH value from 3.25 to 6.25 (Nortoris, et al., 2020). In this study, it was found that the use of lime with a dose of 0.2 g/L can increase the pH value change from 3.31 to 7.01. This value has met the requirements of quality standards, where the pH value of wastewater from good mining activities according to SNI 6989.11:2019 is 6 - 9.

Conclusion

Conclusion Based on the results of the research that has been conducted, several conclusions can be drawn as follows; The combination of zeolite and limestone has proven effective in increasing the pH of Acid Mine Drainage (AMD) from PT. XYZ from an initial value of 2.6 to meet environmental quality standards. The mass ratio between zeolite and limestone significantly affects the rate and extent of pH increase. The mass ratio of 25% zeolite and 75% limestone (V4) is the most optimal composition in this study, which is able to increase the pH to 8.02 in 60 minutes to 8.5 and remained stable to time 90 minutes The presence of zeolite in the mixture is strongly suspected of playing a role in minimizing the effects armouring on limestone particles, thereby increasing the efficiency of the overall neutralization process.

Recommendations

For further development, it is recommended to conduct pilot scale research and analyze the efficiency of reducing dissolved heavy metal levels using instrumentation tools.

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