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Alternative Neutralizers for Acid Mine Wastewater: Characterization and Neutralizing Potency of Pond Ash and Concrete Sludge

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Abstract*— One of the most widely used fuels is coal, where the South Sumatra region is an area with coal potential whose production is still being increased. On the other hand, there are negative impacts caused by coal exploration activities, one of which is mining runoff, commonly called acid mine drainage. Acid mine drainage has a low pH and contains inorganic constituents such as iron and manganese. Therefore, proper and optimum information processing is needed to support environmentally friendly mining management. In contrast, alternative materials such as blast furnace slag, wood ash, fly ash, cement kiln dust, and construction waste are being used. These materials are effective against AMD, at a relatively low cost, and outperform traditional neutralizers. However, comparing them with conventional agents is challenging due to limited data and experimental variation. In this study, the objectives to be achieved are 1) to determine the quality conditions of AMW at the study location, 2) to determine the effectiveness of the use of chemical and alternative compound neutralization agents in the processing process, and 3) to provide recommendations for the dosage, type of neutralization agent, and the most optimum time in AMW processing. The AMW obtained is then subjected to laboratory analysis related to quality, including TSS, pH, Fe, and Mn parameters. In addition, conventional and alternative neutralization agents are also prepared and then used to process AMW. Then, experiments were carried out on variations in the type of neutralization agent, the dosage of neutralization agent use, and the contact time between the neutralization agent and AMW. Experimental results have shown that these materials, mainly pond ash and concrete sludge, can effectively neutralize pH and reduce Mn concentrations by up to 83.26% and 79.12%, respectively. Similarly, Fe concentrations can be reduced by up to 80.76% and 74.05% using pond ash and concrete sludge, respectively. While these results are promising, future research should focus on characterizing the generated sludge to confirm the adsorption of ferrous and manganese ions onto the surface of the alternative neutralizers.

Keywords— concrete sludge, pond ash, neutralizer, acid mine drainage, characterization

I. INTRODUCTION

Coal is a significant fossil fuel source globally. Indonesia ranks third in coal production, with reserves of around 26.2 billion tonnes [1]. In particular, Sumatra accounts for around 10% of Indonesia's coal reserves, making it a significant energy hub [2]. The increasing annual coal production drives mining activities. However, this demand also raises environmental issues, such as acid mine drainage (AMD), characterized by low pH and high concentrations of iron, manganese, and suspended matter [3]. AMD negatively affects aquatic life, soil quality, and human health. Therefore, an important focus is on the efficient treatment of AMD to ensure sustainable coal energy management [1].

Acid mine drainage (AMD) is formed when rocks' sulfide minerals react with water oxygen [2]. Wetland construction is a recommended method for AMD treatment in Indonesia, according to the Decree of the Minister of Environment in 2003. However, the use of this method has limitations: 1) only a few plant species can effectively accumulate metals [3], 2) it requires more time and space [3], and 3) it cannot accumulate high levels of Fe and Mn metals at once [4]. Instead, many Indonesian mining companies use neutralization with chemicals such as calcite lime [5], quicklime (CaO) [6], quicklime (Ca(OH)2) [7], dolomite (CaMg(CO3)2) [8], silica lime (CaSiO3) [9], caustic soda (NaOH) [10], and soda ash (Na2CO3) [5]. This method is preferred because of its efficiency, speed, and space-saving AMD processing. Conventional neutralization for AMD often involves using chemicals classified as Toxic and Hazardous Materials (B3), which can damage the environment. This approach is not sustainable for coal mining.

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In contrast, alternative materials such as blast furnace slag [11], wood ash [12], fly ash [13], cement kiln dust [14], and construction waste [6] are being used. These materials are effective against AMD, at a relatively low cost, and outperform traditional neutralizers. However, comparing them with conventional agents is challenging due to limited data and experimental variation [15], making it difficult for mining companies to select the best neutralization materials and conditions for AMD treatment.

Therefore, this study will comprehensively determine the variation of neutralizing materials, dosage variations, and reaction time variations to determine the most optimum AMD treatment conditions [16]. This optimization process will be carried out using an experimental approach. This study's main objective is to develop intermediate/residue products that can be used as alternative neutralizing materials and develop a prediction model for optimizing AMD treatment. The results of this study can later be utilized by coal mining companies in Sumatra Island and encourage efforts to manage sustainable coal energy.

II. Method

A. Neutralizer Agents

Pond ash, a byproduct generated from the combustion process at PT PLN UPK Tarahan in Lampung Selatan, Indonesia, comprises slag-like particles. These particles vary in size, reaching up to 20 millimeters in diameter. The specific pond ash sample used in this study was sourced directly from the storage warehouse, ensuring its freshness and minimizing any potential alterations to its physical and chemical properties [11]. This fresh sample, which has not undergone drying or prolonged storage, provides a representative sample of the material for analysis and experimentation. Concrete sludge, a waste byproduct generated during the centrifugal production of concrete poles and piles at Radja Readymix in Bandar Lampung, Indonesia, underwent a three-day drying process before sample collection. The sample was extracted from the top layer of the dried sludge and then manually crushed into a fine, powder-like consistency [20]. This careful sample preparation was essential to ensure suitability for subsequent analytical and experimental procedures. By transforming the solid sludge into a powdered form, researchers can more easily conduct various tests, such as chemical analysis, mineralogical characterization, and physical property measurements.

B. Analytical Method

The Agilent Carry 630, CA, USA- FT-IR spectrometer was utilized to acquire Fourier-transform infrared spectra (FT-IR). A powdery sample of FABA and concrete Sludge was compacted into a circular, see-through slice and examined in the transmittance mode

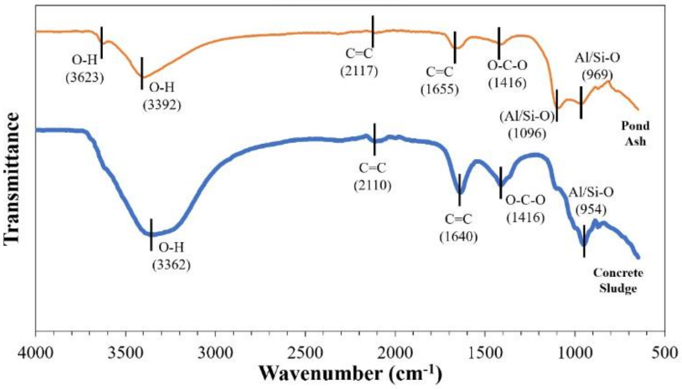


Figure. 1. FTIR results of alternative neutralizer materials: pond ash and concrete sludge

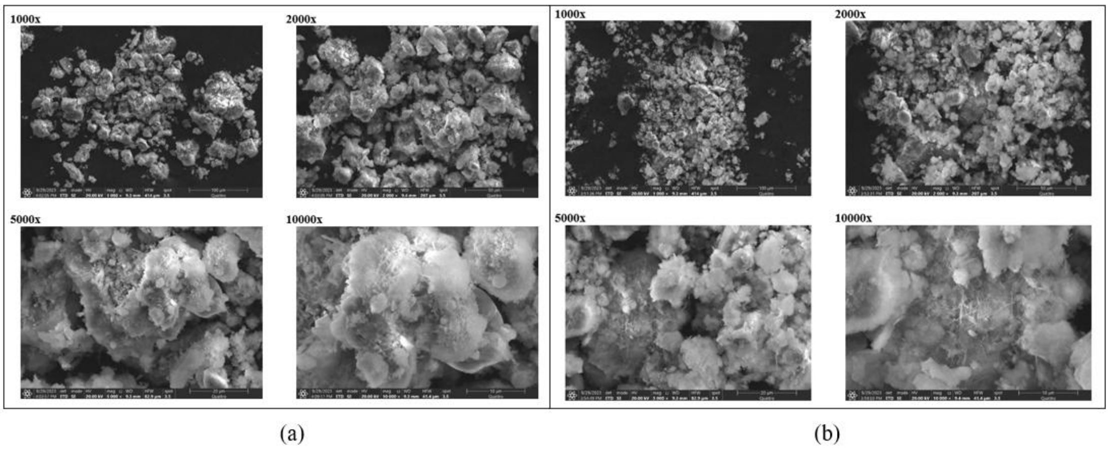


Figure. 2. SEM of (a) Pond ash and (b) Concrete Sludge sample

across a range of wavenumbers, spanning from 400 cm-1 to 700 cm-1 [21]. In the other hand. The chemical compositions and morphologies of these pond ash powders were observed by SEM-EDS Thermo Scientific Quattro with 1000 to 10000x magnification. The pond ash SEM-EDS samples were prepared using the vacuum filtration method proposed earlier. More details can be found in reference [14].

C. Experiments

This experiment aims to evaluate the effectiveness of sodium hydroxide (NaOH) and residual construction mud in neutralizing acid mine drainage and reducing heavy metals Fe and Mn levels. In the initial stage, a certain amount of NaOH (as a strong base) and variations in the mass of construction mud (1-7 grams) were added to the acid mine drainage sample. The mixture was then stirred homogeneously for approximately two hours to ensure an even distribution of chemicals. After mixing, samples were taken and analyzed using an atomic absorption spectrophotometer (HACH DR 1900) to measure the concentration of Fe and Mn. The aim was to determine the extent to which the addition of NaOH and construction mud could reduce the levels of the two heavy metals [22].

In addition, the physical and chemical characteristics of the construction mud that had interacted with the solution were also analyzed using Fourier Transform Infrared Spectroscopy (FTIR) and Scanning Electron Microscope (SEM) techniques. FTIR analysis provides information on the functional groups in the mud sample, while SEM is used to observe the surface morphology and elemental composition qualitatively. Thus, changes in the structure and composition of the mud after the treatment process can be identified [23]. Meanwhile, changes in the solution's pH are also monitored periodically. The pH measurement was conducted before the addition of the reagents (initial pH) and after the stirring process (final pH). Comparing the initial and final pH values will show the effectiveness of NaOH and construction mud in neutralizing the acidity of mine water.

D. Data analysis

After the neutralization experiments were completed, a comprehensive analysis was conducted to evaluate the performance of the most promising conventional and alternative neutralization materials. This analysis encompassed several vital aspects, including the characterization of the neutralizers themselves, the assessment of pollutant concentrations in the treated effluent, and a detailed examination of the quantity and quality of the sludge generated during the neutralization process. By characterizing the neutralizers, we gained valuable insights into their physical and chemical properties, which are crucial for understanding their neutralization mechanisms and optimizing their application. The concentration of pollutants, such as heavy metals and acidity, in the treated effluent, was measured to determine the effectiveness of the neutralization process in reducing pollutant levels [24]. Furthermore, a thorough analysis of the sludge produced during the neutralization process was conducted. This analysis involved quantifying the amount of sludge generated and assessing its physical and chemical properties, including its moisture, pH, and metal content. Understanding the characteristics of the sludge is essential for proper disposal or potential reuse, as it can have significant environmental implications. Through this comprehensive analysis, we aimed to identify the most effective neutralization materials and optimize the neutralization process to achieve the desired water quality standards while minimizing the generation of sludge and its potential environmental impact [25].

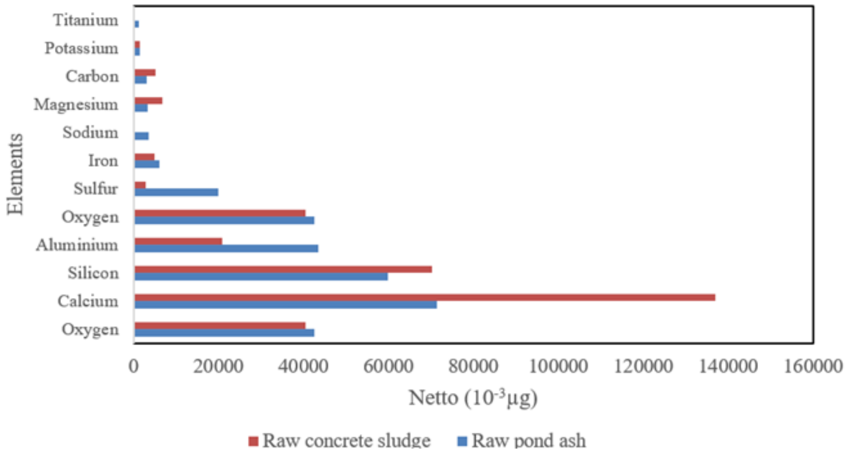


Figure. 3. Elemental compositions of pond ash and concrete sludge

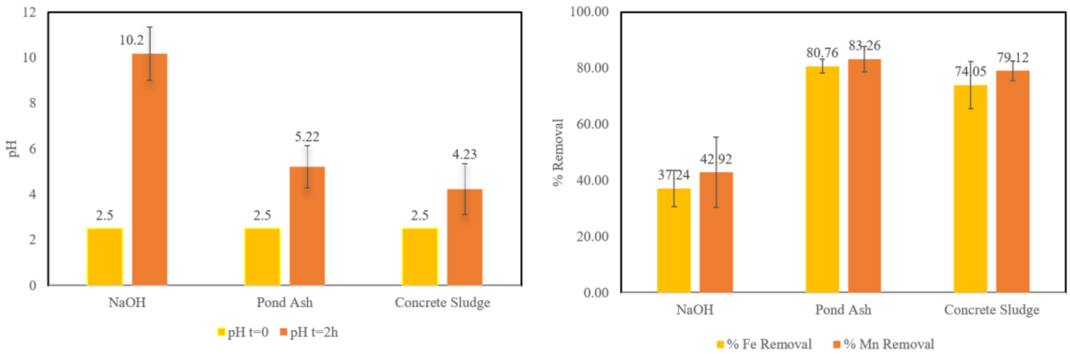
III. Results and Discussion

A. FTIR Analysis

Based on Figure 1, the FTIR results of construction mud show functional groups OH, C=C, CO, C-H, and C - O, which are shown in the wavelength absorption band of 900-4000 cm^{31}. At a wavelength of 3623 cm^{1}, the O-H hydroxyl functional group, that functional group helps clarify water, sugar, and alcohol [23]. The absorption band of 2117.1 cm^{1} shows the C = C alkyne functional group; this is a high carbon content in the wave band [24]. The wave group 1416.4 shows the C - Halkane functional group, while at the C - H alkene functional group, the wavelength is 969.1, and the wavelength of 1095.8 shows the CO functional group [25]. Functional groups are essential characteristics because they can show the nature of carbon.

Meanwhile, based on Figure 1, pond ash has an absorption peak in fly ash bottom ash, indicating the functional group's OH, CC, CO, C-H, and C O shown in the wavelength absorption band of 900-4000 cm^{31}. At a wavelength of 3623 cm^{1}, the O-H hydroxyl functional group, that functional group helps clarify water, sugar, and alcohol [23]. The absorption band of 2117.1 cm shows the CC alkyne functional group; this is a high carbon content in the wave band [24]. At wave 1416.4, it shows the C - Halkane functional group, while in the CH alkene functional group, the wavelength is 969.1, and at a wavelength of 1095.8, it shows the CO functional group [25]. Functional groups are essential characteristics because they can show the nature of carbon.

improvement of various characteristics of soil [15]. In the other hand, the oxigen and calcium content is even more profound in the concrete sludge sample where the calcium detected 136998. 10^{-3} µg. Hydrogen, however, was not detected because its valence electron (1s) does not participate in chemical bonding, and hence, hydrogen is not useful in EDS elemental determination [17]. Oxygen is present in all the materials. Oxygen is responsible for the voids created in the micrographs. The absence of such voids in the SEM images of all sample correlates with the absence of oxygen in the sample [15]. Pond ash, also known as ash pond residue or ash pond waste, refers to the byproduct of coal combustion in thermal power plants. It is a type of coal ash that is collected and stored in large ponds or reservoirs, commonly referred to as ash ponds or ash impoundments. When coal is burned for electricity generation, various solid residues are produced, including fly ash and bottom ash. Fly ash is the fine, powdery material that is carried away with the flue gases, while bottom ash is the heavier, coarser material that settles at the bottom of the combustion chamber. Pond ash is primarily composed of fly ash, along with some bottom ash, that has been collected in the ash pond over time. Pond ash typically contains a mixture of inorganic compounds, including silica, alumina, iron, calcium, and other trace elements present in coal. The composition and characteristics of pond ash can vary depending on the coal source, combustion process, and ash collection methods used at the power plant and its discussed in the SEM Analysis.



(a) (b)

Figure 4. (a) Neutralization performa (b) pollutant removal

B. SEM Analysis

To discuss the relevance between FTIR result and SEM-EDS result, one can compare the chemical composition information obtained from FTIR with the elemental composition information obtained from SEM-EDS. The surface morphology of the particles of the raw material of an alternative neutralizer was examined using a scanning electron microscope (SEM), and the SEM images of the materials viewed at different magnifications are shown in Fig. 2 and Fig. 3. Fig. 2 shows the morphology of the Pond ash sample, which is consistent with previously reported studies [15], [16]. The micrograph appeared to be covered by another surface, and the particles observed were spherical in shape with shiny surfaces that were free of dust. The particles were very fine, with a size of a millimeter or less, and had pores of varying sizes within them [15].

Energy dispersive spectroscopy (EDS) was used to determine the inorganic elements present in all the composite samples. The results obtained are presented in Figure 3. The EDS results show that the main elements in the composites are oxygen and calcium. This agrees with the fact that presented results of laboratory investigation carried on silty sand and pond ash reinforced with randomly distributed polyester fibers. They showed that incorporation of fibers results in the

The typical FTIR spectrum of pond ash is shown in the fig.1, which reveals the three characteristics: bands of silica in the region of 400-4200 1/cm. The sharp band at 475 1/cm is attributed to the bending vibrations o Si-O-Si [31], while the band at 808 cm is due to the symmetric vibrations of Si-O-Si [23] and a broad band at 110: cm is attributed to the asymmetric vibrations of Si-O-Si of silica [22]. The absence of other bands indicates the absence of impurities from the sample. While the comparation with other characteristic spectrum in the fig.2 does show various band, rather i shows saturation which also supports the FTIR data.

Conventional neutralizers (such as Ca (OH)2 and CaCO3) are used extensively in AMD treatment. Ca (OH)2 is one of the most effective and common neutralizers that can neutralize AMD within a relatively short reaction time. *T*he neutralization performance, including identification of main substance that provide alkalinity, removal mechanism of As, Fe, and comparison with conventional neutralizers were confirmed. The sedimentation performance was determined and compared with conventional neutralizers. The high ca*lcium* content of the *material* can be attributed to the styrene monomer's long *O-H* bond [23].

C. AMW neutralization and pollutants removal

Figure 4a compares pH at 0 and 2 hours for three material types: NaOH, Pond Ash, and Concrete Sludge. The initial pH of the three materials is the same, around 2.5. However, after 2 hours, the pH of NaOH increased significantly to around 10.2. Meanwhile, the pH of Pond Ash and Concrete Sludge only increased slightly, to around 5.22 and 4.23, respectively. These results indicate that NaOH can increase the pH of the solution significantly compared to the other two materials.

Figure 4b presents the percentage of Fe and Mn reduction for the same three types of materials. NaOH has the lowest percentage of Fe and Mn reduction, around 37.7% and 42.1%, respectively. In contrast, Pond Ash and Concrete Sludge show higher Fe and Mn reduction percentages. Pond Ash can reduce Fe by 80.76% and Mn by 83.26%, while Concrete Sludge reduces Fe by 79.12% and Mn by 74.05%. From these results, Pond Ash and Concrete Sludge are more effective in lowering Fe and Mn levels than NaOH. Overall, both graphs show that the three materials have different characteristics that influence the pH of the solution and the ability to reduce Fe and Mn levels [25]. NaOH is effective in increasing pH but less effective in reducing heavy metals. At the same time, Pond Ash and Concrete Sludge are more effective in reducing heavy metals but less effective in changing pH.

IV. Conclusion

In this study, each alternative material, characterized by various elements revealed by EDS analysis, demonstrated promising neutralization properties. FTIR analysis confirmed the dominance of hydroxyl (OH) functional groups, particularly from calcium-based compounds, which play a crucial role in the neutralization process. These hydroxyl groups react with metal ions in acid mine wastewater (AMW), forming insoluble metal hydroxides that precipitate out of the solution. Furthermore, SEM micrographs revealed the porous nature of the alternative materials, providing a large surface area that facilitates the adsorption of metal ions. This combined mechanism of precipitation and adsorption contributes significantly to the effectiveness of these alternative materials in reducing pollutant concentrations in AMW. Experimental results have shown that these materials, mainly pond ash and concrete sludge, can effectively neutralize pH and reduce Mn concentrations by up to 83.26% and 79.12%, respectively. Similarly, Fe concentrations can be reduced by up to 80.76% and 74.05% using pond ash and concrete sludge, respectively. While these results are promising, future research should focus on characterizing the generated sludge to confirm the adsorption of ferrous and manganese ions onto the surface of the alternative neutralizers. This will provide further insight into the mechanisms involved and optimize the application of these materials for effective AMW treatment. Developing and using these innovative engineering materials can effectively address the challenges of acid mine wastewater and contribute to sustainable mining practices.

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