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Research Article

Combining biochar with sediment in the treatment for the effectiveness of sulfate and heavy metal Pb reduction of acid mine drainage

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Abstract

The increasing mining activities have led to the problem of acid mine drainage (AMD) pollution. A method that combines biochar treatment as an adsorbent with wetland sediment treatment as a source of sulfate-reducing bacteria is used to address AMD effectively. This research aimed to determine the ability of biochar in combination with wetland sediment treatment to reduce sulfate and heavy metal content in acid mine drainage wastewater. This research was conducted on a laboratory scale in an AMD wastewater treatment reactor with the following treatments of biochar mixed with wetland sediment. Observations included sulfate content, pH, and heavy metal content. Scanning electron microscope (SEM) analysis was also performed on the biochar. SEM observations revealed the presence of small, dense, and irregularly shaped pores on the surface of the biochar. The results on day 30 showed that biochar mixed with wetland sediment was able to reduce sulfate concentration by 74.19% and reduced Pb by 73.79%, compared with treatment sediment only to 64.81% sulfate concentration and reduced Pb by 53.85%, treatment biochar only had reduced sulfate of 46.90% and reduced Pb by 58.67% and control 1.79% sulfate concentration and reduced Pb by 1.87%.

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Introduction

The advancement of the mining industry has had a detrimental impact on the environment due to its waste products. One of the mining wastes is sulfuric acid liquid waste originating from discarded ore residues (Wibowo et al., 2023). This liquid is known as acid mine drainage (AMD), and because of its acidic nature, it can lower the pH of water and dissolve heavy metal ions. AMD is formed from mining excavation sites containing a significant amount of metal sulfides, which, when exposed to air, react with oxygen to form sulfate (SO_4) and, upon contact with water, transform into sulfuric acid (Akcil and Koldas, 2006). Due to its acidic nature, AMD can be lethal to aquatic organisms and disrupt or even kill plant growth on land. Furthermore, AMD serves as a source of heavy metal pollution with health implications for humans (Gaikwad et al., 2011; Du et al., 2022).

One of the heavy metals often found in acid mine drainage is lead (Pb). This heavy metal poses a significant threat to organisms in the environment as it interferes with their metabolism and can cause various human diseases, such as anemia, brain disorders, and nephritis, and is also known to be carcinogenic (Xu et al., 2017). Consequently, the presence of lead in the environment is regulated by the Environmental Protection Agency (EPA) (He et al., 2016). In light of these concerns, it is necessary to address acid mine drainage waste in the environment. Thus far, the treatment of AMD has been carried out chemically using calcium carbonate or physically by burying AMD in large pits to prevent oxygen contact (Muchamad et al., 2021). Both of these methods are highly inefficient, expensive, and environmentally unfriendly. One promising and environmentally friendly alternative is the bio-removal method, which employs sulfate-reducing bacteria (SRB) to reduce sulfate and heavy metals (Fahruddin et al., 2021).

Within wetland sediments, numerous bacteria have the capability to reduce sulfate and heavy metals (Fahruddin et al., 2021). On the other hand, biochar has the ability to serve as a heavy metal adsorbent in acid mine drainage (Wibowo et al., 2023). Therefore, to enhance heavy metal reduction and sulfate reduction further, combining the adsorbent mechanism of biochar with the reduction capability of sulfate-reducing bacteria sourced from wetland sediments is advantageous.

As known, biochar is activated carbon produced through pyrolysis, characterized by fine powder and porosity. It is called biochar because it is made from biomass residues such as rice husks, sugarcane bagasse, wood from forests, coconut shells, and other agricultural waste biomass (Du et al., 2022; Ighalo et al., 2022). Biochar has a pH between 7 and 12 and mainly consists of carbon (40-75%) with small fractions of minerals and volatile organics. Besides, it is somewhat porous due to rudimentary pore drilling and may possess a highly specific surface and a variety of surface functional groups (Fseha et al., 2021). Among these feedstocks, coconut shell is considered the best material for activated carbon production due to its abundant availability, affordability, high microporosity, low ash content, high water solubility, and high reactivity (Al Malki et al., 2023)

According to Yin et al. (2022), the application of biochar is effective because it supports sulfate-reducing bacteria in forming immobile cells during the sulfate-reduction process and maximizes heavy metal adsorption through active carbon ligands. Additionally, biochar, composed of aromatic carbon rings, remains stable in water, can raise pH, and provides nutrients for bacterial growth (Lu et al., 2012; Du et al., 2022). Furthermore, as stated by Wibowo et al. (2023), using biochar is more effective and cost-efficient as an adsorbent in removing heavy metals such as Fe, Mn, Al, Mg, Cu, Zn, Ca, K, Ba, Li, Pb, Ni, and Si from acid mine drainage. From these various studies, it is evident that the high organic matter content in wetland sediments provides an ideal environment for sulfate-reducing bacteria (SRB) to engage in the complex metal reduction processes (Graya and Harding, 2012; Fahruddin et al., 2021). Furthermore, biochar can be classified into various categories, one of the most common being its utilization as a catalyst support for environmental remediation (Ormsby et al., 2012). The study conducted by Yin et al. (2022) found that the use of biochar can lead to a substantial decrease in Cd levels by as much as 40.7%. Similarly, in the study by Lu et al. (2012), the adsorption of Pb^{2+} reached 45-60% using sludge-derived biochar. In another study by Shakya and Tripti (2017), biochar derived from poultry litter pellets was capable of reducing Cu, Zn, and Mn by up to 98%.

The utilization of biochar holds great potential to offer a new solution for improving environmental quality in areas contaminated by heavy metals following mining activities. As discussed earlier, biochars are effective adsorbents for removing contaminants due to their unique properties, including abundant Scope For Growth (SFG). Consequently, biochars have become increasingly important as a remediation solution for pollutants in both industrial and agricultural sectors, contributing to the enhancement of environmental quality (Xiang et al., 2020).

Materials and Methods

Materials

Acid mine drainage was obtained from the Lamuru mining site, Massenrepulu District, Bone Regency. Wetland sediment samples were collected from the Antang Makassar, and coconut shell charcoal was obtained from a traditional market in Makassar.

Initial characterization of sediment and acid mine drainage

Initial characterization of sediment samples included total organic carbon analysis using the TOC method, total nitrogen analysis using the Micro Kjeldahl method, and phosphorus content determination using the Stannous Chloride method (Greenberg, 1989). Characterization of acid mine drainage included sulfate content analysis using a spectrophotometer at a wavelength of 420 nm and pH determination using a pH meter (Greenberg, 1989). The characterization results found that the sediment had a total organic carbon content of 327,000 mg/L, total nitrogen content of 19,200 mg/L, and total phosphorus content of 9.83 mg/L. As for the acid mine drainage, it had a pH of 3.2 and a sulfate content of 1.21 ppm.

The analysis of sediment samples prior to treatment involved determining the levels of organic carbon, total nitrogen, and phosphorus. The acid mine drainage was also characterized by analyzing the sulfate concentration and pH. These three elements play a crucial role in providing nutrients for the growth and development of microbes involved in processing heavy metals such as lead and sulfuric acid mine drainage. The organic matter in the sediment releases hydrogen molecules, which act as electron donors, facilitating microbial activity and enhancing sulfate reduction. This process involves the oxidation of sulfur to sulfites. As for the acid mine drainage with low pH levels and high sulfate concentrations (Pester et al., 2012; Sanchez-Andrea et al., 2012).

Carbonization

Clean and dry coconut shells were chopped into small pieces. A total of 1,000 g of coconut shells were placed in a porcelain crucible and heated in a furnace at 400°C for 1 hour to undergo carbonization. This process produced coconut shell carbon, which was then cooled for activation.

Chemical activation of coconut shell charcoal

Afterward, the material was immersed in a 50 mL HCl solution in 1 L deionized water for 24 hours at room temperature. It was then rinsed with deionized water. Next, an 80 mL KOH solution in 1 L deionized water was used for activation, followed by another 24 hours at room temperature. The mixture was rinsed again and filtered using a Buchner funnel. The charcoal was washed with deionized water until the pH of the filtrate was neutral and then dried. The resulting activated carbon, called biochar, was now ready for use in the treatment process.

Scanning electron microscope (SEM) analysis

The biochar samples produced from coconut shell charcoal were analyzed for their physical properties utilizing a scanning electron microscope (SEM) with a magnification of 5000x. The SEM's exceptional flexibility and extremely high spatial resolution enable the visualization and depiction of surface images on the biochar samples. This investigation aimed to explore the surface morphology of the activated carbon, specifically focusing on the distribution and features of its pores. Chemical activation of coconut shell biochar using a 4% hydrochloric acid solution was conducted, followed by analysis using a scanning electron microscope (SEM) at a magnification of 5000x and a scale of 10 micrometers. This allowed for the observation of the physical characteristics of the activated biochar. The SEM analysis revealed the presence of small, dense, and evenly distributed porous structures on the surface of the activated biochar. These structures play a crucial role in enhancing the binding mechanism of heavy metal pollutants and organic waste. In contrast, the unactivated biochar showed minimal pore formation (Figure 1). The development of these porous morphologies in the activated biochar is anticipated to significantly impact its capacity for metal adsorption and the formation of immobilized cells on its surface. Consequently, the activated biochar is expected to be more efficient in addressing environmental pollution (Wei et al., 2019; Du et al., 2022).

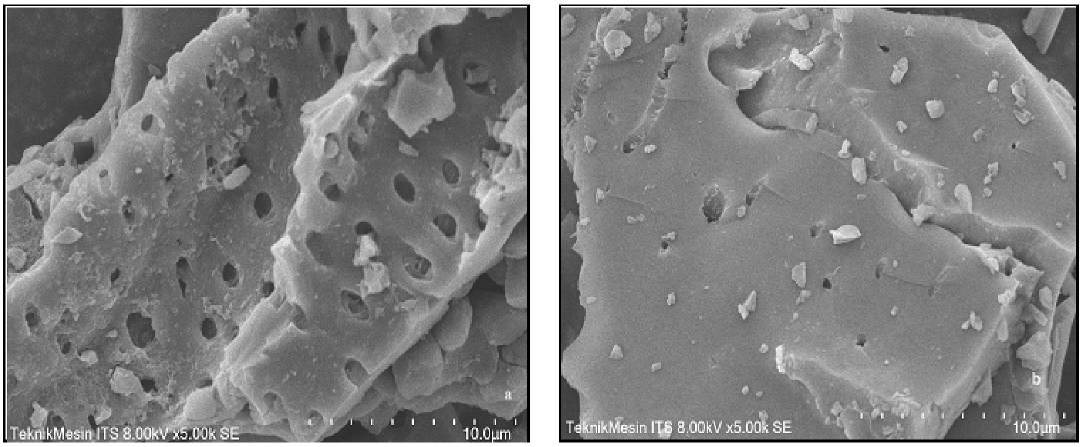

Figure 1. SEM analysis results at 5000x magnification of (a) activated biochar and (b) non-activated biochar.

The activation process releases many volatile compounds, thereby opening pores and reducing hydrocarbon coverage in the biochar. This phenomenon aligns with the findings of Tomczyk et al. (2020), where pore formation and enlargement result from the evaporation of degraded cellulose components, leading to structural differences between activated and non-activated activated carbon pores (Maulina and Mentari, 2018).

Treatment and experiment

The experiments were carried out in the laboratory using modified bioreactor columns based on Fahruddin et al. (2020), measuring 15 cm in diameter and 35 cm in height on a small scale. These columns were filled with 800 ml of acid mine drainage, and the treatments included T1 with 20% sediment and 15% biochar, T2 with 20% sediment, T3 with 15% biochar, and T4 as the control, containing only AMD. Each treatment was replicated and placed at room temperature (27°C). pH and sulfate content observations were recorded on days 0, 5, 10, 15, 20, 25, and 30, while Pb content observations were made on days 0, 10, 20, and 30.

Measurement of pH

pH measurement was conducted using a pH meter, which was calibrated using buffer solutions with pH values of 4 and 7 before stabilization for 15 minutes. Subsequently, the pH meter electrode was immersed in the acid mine drainage sample, and after a brief wait, the pH value was noted based on the pH meter's scale (Greenberg, 1989).

Determination of sulfate concentration

Sulfate concentration was measured using the turbidimetry method by adding barium chloride (BaCl) to the sample solution of AMD. The solution was vigorously shaken using a vortex mixer for 1 min to form colloidal $BaSO_4$, causing the solution to become turbid. The absorbance of the solution was then measured with a spectrophotometer at a wavelength of 420 nm.

Analysis of heavy metal Pb concentration

The analysis of heavy metal Pb began with a sample digestion step. The tested solution was then transferred to a 50 mL measuring flask, rinsed with metal-free water, and added to the flask. Deionized water was introduced to achieve a total sample volume of 25 mL, following which the mixture was homogenized. The next step involved filtration of the solution and preparation for the analysis of heavy metal Pb content through atomic absorption spectroscopy (AAS).

Results and Discussion

The activation process released many volatile compounds, thereby opening pores and reducing hydrocarbon coverage in the biochar. This phenomenon aligns with the findings of Tomczyk et al. (2020), where pore formation and enlargement result from the evaporation of degraded cellulose components, leading to structural differences between activated and non-activated activated carbon pores (Maulina and Mentari, 2018).

pH changes

Based on the pH measurements, all treatments increased pH during the treatment of acid mine drainage (Figure 2). The most notable increase occurred in treatment T1, which involved biochar with sediment, with the pH rising from an initial value of 3.3 to reach pH 6.8 on day 30. Treatment T2, which only received sediment, reached a pH of 6.4. Treatment T3, involving only biochar, resulted in a pH of 5.8, while treatment T4, the control, exhibited a very slight change in pH, reaching pH 3.4 (Figure 2).

Sulfate concentration

The sulfate concentration measurements in the treatment of acid mine drainage revealed the following results: In treatment T1, which involved biochar and sediment, there was a decrease in sulfate concentration from 0.93 ppm, gradually decreasing to 0.24 ppm (74.19%) by day 30. In treatment T2, which only received sediment, the sulfate concentration decreased to 0.38 ppm (64.81%). Treatment T3, involving only biochar, resulted in a sulfate concentration of 0.52 ppm (46.90%) on day 30. In contrast, treatment T4, the control, showed relatively no decrease in sulfate concentration (1.79%) (Figure 3).

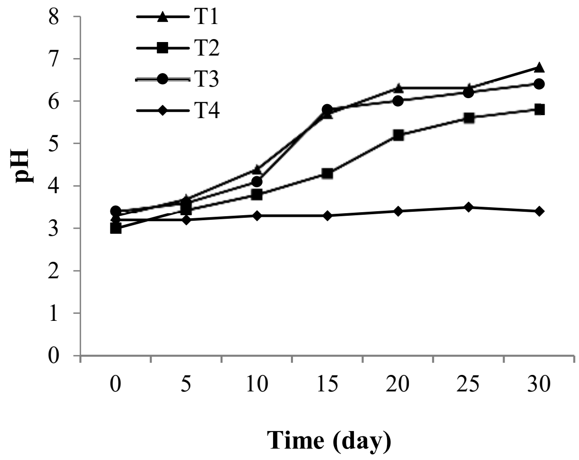

Figure 2. Changes in pH values in the treatment of acid mine drainage, with T1 consisting of biochar and sediment, T2 containing only sediment, T3 including biochar, and T4 as the control.

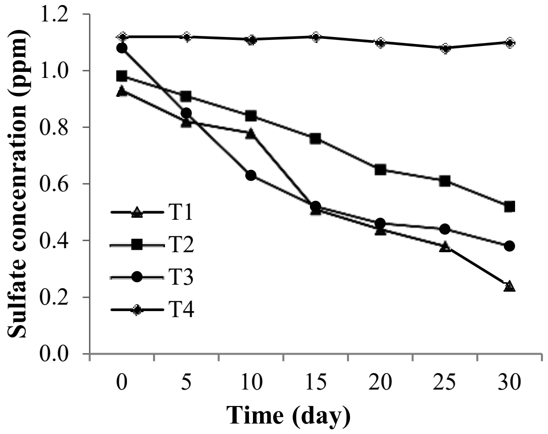

Figure 3. Sulfate concentration in the treatment of acid mine drainage, with T1 consisting of biochar and sediment, T2 containing only sediment, T3 including biochar, and T4 as the control.

Based on both pH and sulfate concentration measurements, it is evident that the increase in pH corresponds to a decrease in sulfate concentration. This indicates that sulfate content is acidic in nature, as lower sulfate concentrations result in higher pH values. The pH levels experienced a significant rise when combining biochar and sediment and treating sediment alone. This increase can be attributed to sulfate-reducing bacteria originating from the sediment. These bacteria reduce sulfate to hydrogen sulfide (HS), decrease sulfate concentration, and produce bicarbonate ions (HCO_3^-) as a buffering agent. Consequently, the pH levels increase within the microenvironment of the acid mine drainage treatment. The formation of bicarbonate indicates the ability of sulfate-reducing bacteria to regulate pH in this experimental setup. Additionally, biochar reduces sulfate concentrations due to its organic material, which acts as an electron donor in the sulfate reduction process. This conversion of sulfate to sulfide contributes to the increase in pH. There was also an increase in pH in the treatment involving only biochar. This can be attributed to releasing alkaline cations, such as Na$^+$, Ca^{2+}, Mg^{2+} and K$^+$, by activated carbon, which forms metal-biochar complexes and raises the pH in acid mine drainage. In contrast, the control treatment did not exhibit a significant increase in pH since there was no sediment or biochar present to participate in sulfate reduction.

The reduction in sulfate concentration observed in the treatment involving the combination of biochar and sediment and the treatment with sediment alone is primarily due to the activity of sulfate-reducing bacteria present in the sediment. These bacteria utilize sulfate as an electron acceptor and the organic matter in the sediment as a carbon source for their metabolism. As a result of sulfate reduction, hydroxide ions (OH$^-$) are released, increasing pH (Pester et al., 2012; Fahruddin et al., 2020). Furthermore, biochar plays a role in decreasing sulfate concentration by serving as an electron donor in sulfate reduction, leading to the conversion of sulfate to sulfide (Du et al., 2022). This is consistent with the research conducted by Yoon et al. (2019) and Lee and Park (2020), indicating that the functional groups on the surface of biochar can trigger active radical species that facilitate the elimination of pollutants via electron transfer or redox reactions.

Heavy metal Pb concentration

The concentration of lead (Pb) was measured during the acid mine drainage treatment, yielding the subsequent outcomes: Treatment T1, which utilized biochar and sediment, exhibited an initial concentration of 1.45 ppm, gradually declining to 0.38 ppm (73.79%) by the 30th day. Likewise, Treatment T2, which solely employed sediment, displayed a lead concentration of 0.72 ppm (53.85%) on day 30. Treatment T3, involving only biochar, resulted in a lead concentration of 0.62 ppm (58.67%) on day 30. Conversely, Treatment T4, the control, demonstrated a relatively minor reduction, with the initial concentration of 1.59 ppm decreasing to 1.57 ppm (1.87%) (Figure 4).

The decline in Pb concentration in the acid mine drainage treatment can be due to biological processes, particularly the involvement of sulfate-reducing bacteria originating from the sediment. These bacteria are accountable for decreasing heavy metal Pb (Fahruddin et al., 2021; Soda and Nguyen, 2023). Another biological process includes the development of immobilized bacterial cells on biochar, which aids in the bioabsorption of heavy metals. Furthermore, the decrease in heavy metal Pb concentration is impacted by chemical processes taking place on biochar, where adsorbent mechanisms are involved in active carbon biochar (Lee and Park, 2020; Xiang et al., 2020).

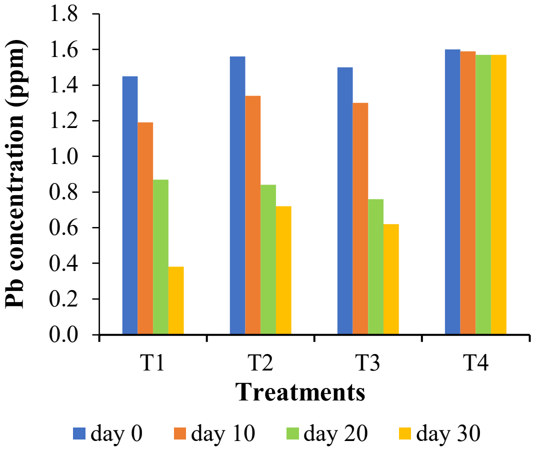

Figure 4. Pb concentration in the treatment of acid mine drainage, with T1 consisting of biochar and sediment, T2 containing only sediment, T3 including biochar, and T4 as the control.

The function of sulfate-reducing bacteria present in the sediment in reducing heavy metal Pb is accomplished through sulfate reactions, leading to hydrogen sulfide (H_2S) production. This newly formed hydrogen sulfide is highly reactive, causing swift reactions with metals and forming insoluble metal sulfide compounds. As a result, some heavy metals precipitate out, while others are attached to immobilized cells and biochar (Khan et al., 2020; Zhang et al., 2023). When biochar was used as the only treatment, a decrease in lead (Pb) concentration was observed. This decrease is attributed to the porous nature of biochar, which acts as an adsorbent for heavy metals like lead, aiding in their absorption and binding (Maulina and Mentari, 2018; Wei et al., 2019).

The adsorption capacity is improved by reducing the particle size of biochar, leading to a larger surface area that can adsorb more ions. In contrast, the control did not show a significant decrease in lead (Pb) concentration because it lacked sulfate-reducing bacteria and activated carbon, which are necessary for binding heavy metal Pb (Tomczyk et al., 2020; Du et al., 2022; Al Malki et al., 2023). The comparison of the efficacy in reducing sulfate and heavy metal Pb between biochar and sediment reveals that both materials are essential in the reduction process. Treatment T2, utilizing sediment alone, exhibited a more pronounced impact on sulfate reduction, resulting in a 64.81% decrease in sulfate concentration and an increase in pH. In contrast, treatment T3, which solely utilized biochar, reduced sulfate levels by 46.90%. However, treatment T3 proved to be more effective in reducing the concentration of heavy metal Pb, achieving a 58.67% reduction compared to T2 53.85% reduction. This indicates that sulfate-reducing bacteria primarily contribute to sulfate reduction by converting it to sulfite, while biochar is more involved in removing heavy metal Pb through adsorption mechanisms. Hence, the combination of biochar and sediment is crucial for the efficient treatment of acid mine drainage.

Combining biochar with sediment has proven to be more effective in terms of reducing sulfate and heavy metal Pb concentrations. Biochar plays a crucial role in mitigating pollutant contamination in the environment, particularly in cases of acid mine drainage. According to a study by Zhang et al. (2023), biochar can enhance the binding of heavy metals to sediment organic matter. Furthermore, biochar is a carrier for bacterial growth and immobilization, facilitating the absorption of heavy metals by immobilized cells. This finding aligns with the statement by Zhang et al. (2022), which suggests that biochar facilitates electron transfer between microbial cells and contaminants resulting from microbe-biochar interactions with sediment organic carbon. Another study by Xu et al. (2017) highlights that numerous bacterial species, particularly *Pseudomonas* sp., can act as adsorbents for heavy metals through the immobilization of Pb.

Considering the discussion above, it is evident that biochar serves as an effective adsorbent for mitigating organic and heavy metal contaminants through various mechanisms. These mechanisms include electrostatic attraction, polar and non-polar organic attraction to the biochar carbonization phase, and partitioning to the non-carbonization phase. Therefore, biochar is vital in interacting with contaminants (Xiaomin et al., 2017).

Conclusion

Utilizing biochar alongside wetland sediment treatment has proven to be highly efficient in mitigating sulfate and heavy metals in acid mine drainage. Biochar is an adsorbent, whereas wetland sediment is a reservoir of sulfate-reducing bacteria inoculum. These two methods work synergistically to create immobilized cells that reduce sulfates and heavy metals in acid mine drainage. The findings of this study demonstrate that the combined use of biochar and sediment is particularly effective in decreasing sulfate levels, resulting in a reduction of 74.19% while simultaneously increasing the pH from 3.3 to 6.8. Similarly, it demonstrates significant efficiency in reducing the concentration of heavy metal Pb by 73.79% compared to using only biochar (58.67%) or only sediment (53.85%).

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