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Research Article

Management of mine acid drainage in a constructed wetland using hyacinth plant and addition of organic materials

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Abstract: Coal mining is an activity to exploit land to get coal stored underground. Environmental problems in coal mining activities are generally associated with Acid Mine Drainage (AMD). The purpose of this study was to find out the best combination of organic matter and incubation time in remediating acid mine drainage on wetlands with water hyacinth plants. This research was conducted at PT Berau Coal in Tanjung Redeb, Berau, East Kalimantan. This study used a split-plot design. The main plot was the treatment of a combination of organic matter which consisted of 4 levels: A0 as a control (without organic matter), A1 with a combination of compost + sawdust (1:2), A2 with a combination of compost + sawdust (1:1), A3 with a combination of compost + sawdust (2:1). The subplot was treatment incubation time consisting of 2 levels, namely for 15 days and 33 days. Data analysis used variance analysis at the 5% level followed by the Duncan Multiple Range Test with a level of 5% when there were significant differences. The results showed that the addition of organic matter had a significant effect on the increase in pH and a decrease in the concentration of Mn in water, but it did not significantly affect the decrease in Fe concentration in water. The best combination of organic matter in acid mine remediation in this study was compost + sawdust (2:1) during the incubation time of 33 days.

Keywords: *acid mine drainage, hyacinth plants, organic matter, remediation, wetlands*

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Introduction

Indonesia is a country that is rich in mining materials, one of which is coal. Coal mining is an activity to exploit land to get coal stored underground. Coal mining companies in Indonesia are very numerous, one of which is PT Berau Coal in Berau, East Kalimantan. Coal mining is generally carried out by open pit mining so that it will have an impact on changes in the landscape, physical, chemical and biological properties of the soil. This impact will automatically disrupt the ecosystem above it, namely the water system.

Environmental problems due to coal mining activities are generally related to Acid Mine Drainage (AMD). The water is formed as a result of oxidation of certain sulfide minerals contained in rocks by oxygen in the air an aqueous environment. Sulfide minerals that are often found in coal mining, namely pyrite minerals (FeS_2). The acid mine drainage has a low pH of 1.5 until 4 and a high content of heavy metals such as Fe >100 mg/l (Nasir, 2014). In mining, many heavy metals are produced and are certainly harmful to the environment, so it will damage the environment if the acid mine drainage is immediately disposed of without management.

The acid mine drainage management technology that is often used is active management, namely by adding lime. However, this is an obstacle for companies because of the high costs required. Another technology is passive acid mine management, one of which is constructed wetland. In this wetland system, it only requires a relatively low cost and does not require complicated maintenance, but the problem is a fairly long remediation time compared to the active management. In anaerobic wetland systems, the composition of the matrix used such as organic matter added with activated sludge can stimulate the growth of sulfate-reducing bacteria to increase alkalinity which can increase AMD pH (Chang et al., 2000).

One of the water plants that has been studied effectively to remediate acid mine drainage is water hyacinth plants. Water hyacinth can absorb heavy metals iron (Fe) and manganese (Mn). Madaniyah (2016) stated that the treatment with water hyacinth plants in wetland ponds decreased dissolved Fe and Mn levels in wetland ponds. Remediation of acid mine drainage can also be done by adding organic matter. According to research by Riwandi and Munawar (2007), organic matter can be used to remediate acid mine drainage. This is because adding organic matter can increase the pH of acid mine drainage and reduce dissolved heavy metals. Remediation of acid mine drainage with bark, compost, and sawdust gives the best results in remediating acid mine drainage.

There have been many studies which state that water hyacinth plants and organic matter can be used to remediate acid mine drainage. In this study, a combination of organic matter was made as a base for constructed wetlands to be planted with water hyacinth plants so that the process of neutralizing mine acid water can take place quickly and effectively.

Material and Methods

The research was conducted at Sambarata Site, PT Berau Coal, Tanjung Redeb, Berau, East Kalimantan from July to September 2018. Analysis of acid mine drainage samples was carried out at PT Berau Coal Site Lati Laboratory, while the analyses of sediment and plant samples were carried out at Sucofindo Cibitung Laboratory.

Acid mine and sludges were obtained from the Water Monitoring Point (WMP) settling pond 36 PIT C2C Site Sambarata PT. Berau Coal. The plant used was water hyacinth (*Eichhornia crassipes*) obtained from Segah River, Berau, East Kalimantan. Organic materials used were compost of cow manure and sawdust. The compost of cow manure was obtained from the composting project at PT Berau Coal Site Sambarata, while the sawdust was obtained from bengkirai wood sawmills nearby.

Preliminary analysis

The acid mine drainage used was previously analyzed for pH, iron (Fe), and manganese (Mn) levels. Results of the analysis are presented in Table 1. Water hyacinth plant was analyzed for iron (Fe) and manganese (Mn) contents. For the organic matter, analyses of C/N ratio, iron (Fe) and manganese (Mn) contents were carried out. The C/N analysis of organic matter was aimed to determine the maturity of organic matter based on the ratio of carbon and nitrogen contained in compost and sawdust. Results of organic matter analysis were presented in Table 2.

Table 1. Characteristics of mine acid sediment pond WMP 36 ST.

Parameters	Sediment Pond	Maximum Value*)
pH	2.85	6-9
Fe total (mg/l)	6.03	7
Mn total (mg/l)	6.68	4

*) Environmental Quality Standards of East Kalimantan Provincial Regulation No. 2 of 2011 concerning Management of Water Quality and Water Pollution Control.

Based on the data in Table 1, it can be explained that acid mine drainage is mine water that does not or contains a little alkalinity (pH <4.5) and contains Fe, Al, Mn, and other metals, high concentrations of acid (H^+) (Skousen, 1996). The high acidity of water with pH 2.85 causes high dissolved metal concentrations. When compared with the environmental quality standards of the East Kalimantan Provincial Regulation No. 2 of 2011 concerning Management of Water Quality and Water Pollution Control, the acidic water does not meet the quality standard, because the pH is too low and the manganese (Mn) level is still above the standard. Therefore, it is necessary to conduct acid mine drainage treatment before being discharged into the river so that the water does not damage the environment.

Table 2. Characteristics of organic materials used for the study.

Materials	Fe (ppm)	Mn (ppm)	C/N Ratio
Compost	0.05	0.04	15.2
Sawdust	0.99	0.26	238

Plant acclimatization

Before water hyacinth plants were planted in constructed wetlands, the plants were acclimatized by planting them in acid mine drainage having pH that was close to pH in the constructed wetlands. The acclimatization site was at Water Monitoring Point (WMP) 12 with a pH of 4 for 7 days. This was aimed to ensure the viability of water hyacinth at acidic pH.

The implementation of research

This research was carried out with a static system. The acid mine drainage which was accommodated in the pond was not drained but only remediated with the material in the reservoir. The constructed wetland was made of 80x80x60 cm^3 size (Figure 1), so that the AMD volume capacity was 270 litres/tank, with a water level of ± 42 cm. The type of wetland pond used was an anaerobic wetland.

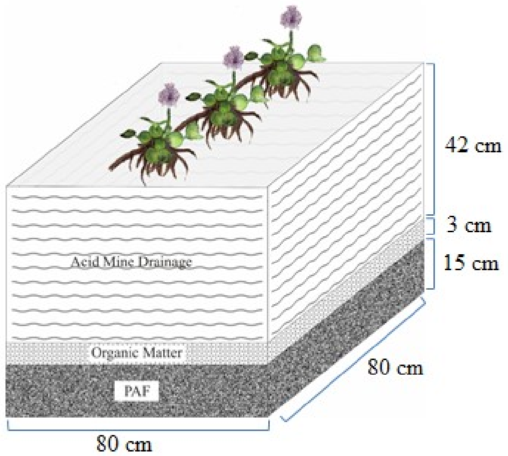

Figure 1. Constructed wetland design.

Sludge having the potential to form an acid (Potential Acid Forming (PAF)) was put into the pond until reaching a height of 15 cm. The next component was an organic matter which was a combination of compost cow manure and sawdust. The organic matter was added until reaching a height of 3 cm. AMD was included in the experimental pond with a water level of ± 42 cm, then 15 clumps water hyacinth plants were added. Plants used for treatment were those having relatively the same weight of 30 grams. This research used a split-plot design. The main plot was the treatment of a combination organic matter consisting of 4 levels, namely A0 without the organic matter, A1 with a combination of compost + sawdust (1:2), A2 with compost + sawdust (1:1), and A3 with compost + sawdust (2:1). The subplot was the treatment of incubation time consisting of 2 levels, namely T1 with a 15 days incubation time and T2 with a 33 days incubation time.

Results and Discussion

Effect of organic material composition on pH

There was a gradual increase in pH from the original 2.85 to 4.15 (Table 3). As with the control treatment without organic matter (A0), the results of pH ranged from 3.02 to 3.05.

Table 3. Average pH of a water.

Treatment	A0	A1	A2	A3	Average
T1	3.05p (c)	3.42q (b)	3.50p (b)	3.90q (a)	3.47k
T2	3.02p (c)	3.63p (b)	3.49p (b)	4.15p (a)	3.57 j
Average	3.04z	3.53y	3.50y	4.03x	+

Description: The mean followed by the same letter in the row and column shows that there is no significant difference based on the DMRT test at the level of 5%. The sign (+) indicates an interaction.

Data presented in Table 3 show that there was an interaction between the treatment of a combination of organic matter and the treatment of incubation time. The treatments of a combination of organic matter A0 and A2 in the treatments of incubation time of T1 and T2 were not significantly different, but the treatments of a combination organic materials A1 and A3 in the treatments of incubation time T1 and T2 were significantly different. While for the treatment of the incubation time of T1 in all treatments the combination of organic materials both A0, A1, A2, and A3 were significantly different, as well as for the incubation time of T2. The treatment that gave the optimum effect was the combination of A3 organic matter namely compost + sawdust (2: 1) and an incubation time of T2 (33 days). This shows that the adding of organic matter increased the pH of the acid mine drainage. At the 33rd day, the results were higher than that of the 15th day. This means that the longer remediation process in constructed wetlands will increase the pH of the water too. The increase in pH can be due to an inundated condition where there is a reduction process where Fe^{3+} ions will be reduced to Fe^{2+} which releases OH^- ions which will bind H^+ ions so that the acidity decreases and the pH increases (Stevenson, 1982). Compost is a mature organic material that contains many organic acids. This organic acid is used by sulphate-reducing bacteria as electron donors, so the process of reducing sulphates can occur faster and pH can rise better. The more compost composition used, the higher the pH of the water will be. Sulphate-reducing bacteria are characterized by anaerobic respiration using sulphate as the electron receiving centre. Sulphate reduction reactions are as follow:

$$2C_2H_5OH + SO_4^{2-} \rightarrow 2CH_3COOH + S^{2-} + 2H_2O$$

Sulphate reduction is the main cause in pH neutralization and reduction of toxic sulphates and metals. Sulphate-reducing bacteria produce 2 moles of alkalinity per one mole of sulphate which is reduced. The role of organic material is as a producer of alkalinity (bicarbonate), where acetate as an electron donor and sulphate-reducing bacteria as the main actors in the process of reducing sulphate. The alkalinity reaction can be seen in the following reaction:

$$2CH_3COO^- + SO_4^{2-} + H^+ \rightarrow H_2S + 2HCO_3^-$$

The A0 treatment showed stable results of pH ranging from 3.02 - 3.05. This is because the solid media of constructed wetlands used in this study did not contain organic matter which could stimulate the growth of sulphate-reducing bacteria.

Effect of constructed wetlands on Fe and Mn

The average Fe concentration in wetland media water is presented in Table 4. The results of this study showed that all treatments gave results that were not significantly different. In constructed wetland have been able to reduce dissolved Fe concentrations in acid mine drainage. The decrease in metal availability also occurs chemically due to the increase in pH values that occur due to the addition of organic matter and sulphate-reducing bacteria activity. This happens because the activity of microorganisms in wetlands produces H_2S that is reactive and immediately reacts with metals to form metal compounds that are difficult to dissolve (Prianto, 2016). As a result of the activity of these microorganisms, the metal will be precipitated so that the metal solubility becomes low and the concentration is expected to be not harmful to the environment. There are four possibility processes of decreasing heavy metals, namely (1) the interaction between sulphides (S^{2-}) produced in the process of reducing sulphates with valentic metals 2 (such as Fe^{2+}) forming precipitated metal sulphides, (2) the process of metal absorption by plant tissue, (3) the process of metal adsorption by organic matter, and (4) the process of metal biopsy by microorganisms found in wetland environments (Madaniyah, 2016). The other parameter measured in this study was dissolved of Mn. The average Mn concentrations after incubation 15 and 33 days are shown in Table 5. Data of the treatment of T2 or 33 days, showed that the higher the compost combination used, so the lower the metal solubility of Mn (Table 5). The interaction between the two factors showed that the treatments of the combination of organic matter A1, A2 and A3 in the treatments of incubation time of T1 and T2 were not significantly different, but the treatment of the combination of organic matter A0 in the treatment of incubation time T1 and T2 was significantly different.

Table 4. Average Fe concentration in water (mg/l).

Treatment	A0	A1	A2	A3	Average
T1	12.58	0.65	0.44	0.56	3.56j
T2	7.53	2.44	3.06	4.03	4.27j
Average	10.06x	1.55x	1.75x	2.29x	-

Description: The mean followed by the same letter in the row and column shows that there is no significant difference based on the DMRT test at the level of 5%.

Table 5. Average Mn concentration in water (mg/l).

Treatment	A0	A1	A2	A3	Average
T1	13.45q (a)	17.24p (a)	12.99p (a)	15.62p (a)	14.83k
T2	21.65p (a)	18.92p (ab)	15.46p (b)	14.44p (b)	17.62 j
Average	17.55x	18.08 x	14.23 y	15.03 xy	+

Description: The mean followed by the same letter in the row and column shows that there is no significant difference based on the DMRT test at the level of 5%. The sign (+) indicates an interaction.

Whereas for the treatment of incubation time T1 in all treatments, the combination of organic materials both A0, A1, A2, and A3 were not significantly different, but for treatment the incubation time of T2 the A0 was significantly different from A2 and A3 but was not significantly different from A1 and A1 was not significantly different with A2 and A3. The treatment that gave the optimum effect was the combination of A3 organic matter namely compost + sawdust (2: 1) and an incubation time of T2 (33 days). This is because compost is a mature organic material containing humus. Prasetiyono (2015) found that compost can be used to minimize heavy metals because compost contains humic acid and fulvic acid that can adsorb heavy metals by the functional groups in both substances. These humus substances contain a group of carboxyl or -COOH functions. In compost that has been decomposed, the functional group will undergo a deprotonization process so that the H + ion will escape from its compound and the negatively charged functional group. The pattern of removal of heavy metals by humus is by adsorbing metal ions and also forming complex compounds and chelates so that the metal is difficult to free. This chelation process can be caused by an inter-load bond called an electrostatic chelate. This is what happens in the binding of heavy metals Fe and Mn by organic matter. Nurhayati and Sutrisno (2011) stated that compost containing humus substance (fulvic acid, humic acid and humin) was able to adsorb heavy metal complexes through cation exchange, chelate formation, and electrostatic bonds. When compared with dissolved Mn concentrations before treatment, the results of the study showed an increase in dissolved Mn concentration. This can be caused by the reduction of MnO_2 found in sludge to Mn^{2+}. The solubility of Mn^{2+} becomes high in the water so that when the results are analyzed the results are greater. Because when conditions are inundated (reduction), Mn will be reduced earlier than Fe so that Mn^{2+} is greater than Fe^{2+}. The reaction of manganese reduction occurs as follows:

$$MnO_2 + 4H^+ + 2e^- \longrightarrow Mn^{2+} + H_2O$$
$$Mn_2O_3 + 6H^+ + 2e^- \longrightarrow 2Mn^{2+} + 3H_2O$$
$$Mn_3O_4 + 3H^+ + 2e^- \longrightarrow 3Mn^{2+} + 4H_2O$$

Accumulation of Fe and Mn in sediment

The A0 treatment did not use organic matter, treatment A1 with a combination of organic compost + sawdust (1:2), treatment A2 with compost + sawdust (1:1), and treatment A3 with compost + sawdust (2:1). Low compost C/N ratio shows that the compost used is ripe, as is the case with sawdust with a very high C/N ratio. From the treatment, the lowest C/N ratio is in treatment A3 with a combination of compost + sawdust (2:1). In addition, Fe and Mn levels in organic matter are also quite low. The results of Fe and Mn levels in sediments can be seen in Table 6 and Table 7, respectively. The A0 treatment showed significant differences from the other treatments. While for A1, A2, and A3 treatments showed no significant difference from each other. The incubation time showed that they were not significantly different. This proves that organic material can absorb Fe and Mn in large concentrations for a long time. The A0 treatment showed the highest Fe and Mn concentration because the sludge used was pure from the WMP 36 sediment pond without mixed organic matter so that it was highly acidic with very high Fe and Mn content. This was indicated by the colour of blackish sludge with yellow boy deposits.

Table 6. Average Fe concentration in sediments (ppm).

Treatment	A0	A1	A2	A3	Average
T1	383.7	179.67	117.67	119.33	200.08j
T2	347.3	184.67	304	259	273.75j
Average	365.5x	182.17y	210.83y	189.17y	-

Description: The mean followed by the same letter in the row and column shows that there is no significant difference based on the DMRT test at the level of 5%.

Table 7. Average Mn concentration in sediment (ppm).

Treatment	A0	A1	A2	A3	Average
T1	153.3	71.33	46.67	47.33	79.67j
T2	138.7	73.33	121.3	103.33	109.17j
Average	146x	72.33y	84y	75.33y	-

Description: The mean followed by the same letter in the row and column shows that there is no significant difference based on the DMRT test at the level of 5%.

The role of organic materials is as follow:

$$2CH_3COO^- + SO_4^{2-} + H^+ \longrightarrow H_2S + 2HCO_3^-$$

One mole of sulphide will be produced from one mole of sulphate in the process of reducing sulphate, this sulphide will precipitate heavy metals by forming metal soluble with low solubility.

$$Me^{2+} + H_2S \longrightarrow MeS + 2H^+$$

Absorption of metal ions by non-living organisms is believed to occur through an adsorption process that involves functional groups related to proteins, polysaccharides, carboxylates, hydroxyls, sulfhydryl groups and other biopolymers found in cells or cell walls (Intan, 2016). These polar groups are thought to be able to interact with heavy metals. The absorption mechanism that occurs between –OH groups that the groups are bound to the surface with positively charged metal ions (cations) is an ion exchange mechanism (Nurhayati and Sutrisno, 2011). The levels of Fe and Mn in the A1, A2, and A3 treatments showed smaller results compared to the A0. This was due to a large number of Fe and Mn elements that can be absorbed by organic matter. The pattern of removal of heavy metals by organic matter is by adsorbing metal ions and also forming complex compounds and chelates so that the metal is difficult to lose.

Accumulation of Fe and Mn in hyacinth plant

The results of Fe accumulation in plant tissue can be seen in Table 8. The root tissue of the water hyacinth plant before treatment showed no Fe concentration. Data in Table 8 present that in all treatments, water hyacinth plants could absorb heavy metals Fe with different concentrations. The highest Fe content in the water hyacinth root was in the A0 treatment that was significantly different from that in the A1 and A3 treatments, but it was not significantly different from the A2 treatment. The second factor (the incubation time) did not show a significantly different result. This shows that both at day 15 and day 33 of the water hyacinth plants had not experienced saturation in absorbing heavy metals. Mn levels in water hyacinth plants presented Table 9 indicated that Mn in the root tissue of the water hyacinth plant before the treatment was not detected. The interaction between the two factors showed that the treatments of the combination of organic materials of A0, A1, A2, and A3 in the treatments of incubation time of T1 and T2 were all significantly different. The treatment that gave the optimum effect was the A1 treatment is without the addition of organic matter and incubation time of T2 (33 days).

Table 8. Average Fe concentration in water hyacinth roots (ppm).

Treatment	A0	A1	A2	A3	Average
T1	0.29	0.12	0.29	0.18	0.22j
T2	0.45	0.15	0.19	0.24	0.26j
Average	0.37x	0.13y	0.24xy	0.21y	-

Description: The mean followed by the same letter in the row and column shows that there is no significant difference based on the DMRT test at the level of 5%.

Table 9. Average Mn concentration in water hyacinth roots (ppm)

Treatment	A0	A1	A2	A3	Average
T1	0.14q (a)	0.17q (a)	0.13q (a)	0.16q (a)	0.15k
T2	0.43p (a)	0.38p (ab)	0.31p (bc)	0.29p (c)	0.35j
Average	0.29x	0.28x	0.22y	0.22y	+

Description: The mean followed by the same letter in the row and column shows that there is no significant difference based on the DMRT test at the level of 5%. The sign (+) indicates an interaction.

Observations from day 15 and day 33 showed the increase in Mn concentration in water hyacinth plants. This could be due to the increase the dissolved Mn in acid mine drainage, so this was directly proportional to the absorption of Mn content by the roots of the water hyacinth plant. From the two parameters above, the highest levels of Fe and Mn in the water hyacinth root were in the A0 treatment. This happened because there was no organic material in treatment A0 so that the plants were the only object that could absorb metals, thus showing the levels of Fe and Mn in tall plants. If seen from the biomass of water hyacinth plants, in the A0 treatment showed a small amount of biomass because most of the water hyacinth plants died. This shows that if without organic matter, water hyacinth plants can absorb Mn metal but cannot long lasting and eventually die. Another case in other treatments, water hyacinth plants can grow well and even have multiplied by producing saplings. Metal accumulation by plants depends on many factors, namely (1) the nature of plants, such as: species, growth speed, size and depth of roots, a speed of evaporation, and nutrient requirements for metabolism, (2) soil factors, such as: pH, content and nature of organic matter, nutrient status, number of metal ions and certain anions, dan (3) environmental and management variables namely temperature, humidity, sunlight, rainfall, fertilization and others (Irhamni, 2017). The next parameter calculated is the absorption of Fe and Mn levels by water hyacinth plants in Table 10.

Table 10. Results of calculation of Fe and Mn absorption in water hyacinth roots.

Treatment	A0	A1	A2	A3
Dry weight (gram)	2.9	3.4	3.7	4.1
Fe concentration in plants (ppm)	0.37	0.13	0.24	0.21
Fe uptake by plants (gram/clump)	1.07	0.44	0.89	0.86
Mn concentration in plants (ppm)	0.29	0.28	0.22	0.22
Fe uptake by plants (gram/clump)	0.84	0.95	0.81	0.90

According to Kelly (1997), the mechanism of action of phytoremediation may include (1) phytoextraction that is the absorption of pollutants by plants from water or soil and then accumulated or stored in plants (leaves or stems), such plants are called hyperaccumulators, (2) rhizofiltration that is the utilization of the ability of the roots to absorb, precipitate, and accumulate heavy metals. But usually this basic concept applies if the media that is contaminated is a body of water, (3) phytodegradation that is the process of absorption of pollutants by plants and then these pollutants undergo metabolism in plants; the pollutant metabolism in plants involves enzymes including nitroreductase, laccase, dehalogenase and nitrilase, (4) phytostabilization is a process carried out by plants to transform pollutants in the soil into non-toxic compounds without first absorbing these pollutants into the plant's body, and (5) phytovolatilization that is the process of absorption of pollutants by plants and these pollutants are changed to be volatile and then transmitted by plants; the pollutants released by plants into the air can be the same as the initial form of pollutant compounds, can also be different compounds from the initial compound.

Conclusion

Constructed wetland systems increased the pH of acid mine drainage and reduced the solubility of heavy metals Fe and Mn. The addition of organic matter had a significant effect on the increase in pH and a decrease in dissolved Mn concentration in the remediation of acid mine drainage, but had no significant effect on the decrease in Fe concentration in water. Organic materials were also able to absorb heavy metals Fe and Mn. The composition of the best organic matter in acid mine remediation in this study was in the A3 treatment, namely with compost + sawdust (2:1) at 33 days incubation time. Water hyacinth plants could absorb heavy metals Fe and Mn in artificial wetland systems.

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