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Effectivity of *Lepironia articulate* Retz (Domin.) As an Absorbend of Heavy Metals Fe and Mn Acid Mine Drainage

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Abstract

Indonesia is one of the world's largest coal-producing countries. Most coal mining activities are conducted using the open-pit method. A classic problem caused by mining activities is acid mine drainage (AMD). Various conventional methods have been developed to increase the pH of the water and remove heavy metals such as iron (Fe) and manganese (Mn) in acid mine drainage, including chemical precipi-tation, co-agulation, flocculation, microorganisms, adsorption, and passive treatment methods. In this study, a pas-sive treatment method using constructed wetlands with phytoremediation techniques using L. articulate plants was employed. This study aims to analyze the effectiveness of the L. articulate plant in increasing the acidity of water and reducing the levels of Fe and Mn metals dissolved in acid mine drainage. The re-sults of the initial sample analysis of acid mine drainage showed a pH of 2.8. The initial values of Fe met-al concentrations were 0.09 mg/L and Mn 8.10 mg/L. After the phytoremediation process, it was found that L. articulate plants were effective in increasing the pH value and reducing the concentrations of dis-solved Fe and Mn metals in acid mine drainage within 20 days. The percentage reduction was 93.33% in the organic reactor and 55.56% in the inorganic reactor for Fe metal concentrations. For Mn metal reduc-tion, the percentages were 38.02% in the organic reactor and 25.80% in the inorganic reactor.

Keywords : Phytoremediation, Lapironia articulate, constructed wetland, Acid Mine Drainage, Iron, Manganese Metals.

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1. Introduction

The classic problem caused by coal mining activities is acid mine drainage (AMD). Acid mine drainage is formed when ore materials containing oxidized sulfide minerals (such as pyrite) mix with water[1]. The solubility level of acid mine drainage can erode rocks and soil, leading to the dissolution of various elements such as iron (Fe), manganese (Mn), cadmium (Cd), and zinc (Zn)[2]. In its processing, acid mine drainage is regulated by the Ministry of Environment Regulation No. 5 of 2023 on "Quality Standards for Wastewater Management for Coal Mining Enterprises and/or Activities." Additionally, it is also gov-erned by the South Sumatra Governor Regulation No. 12 of 2012 on "River Water Quality Standards."

A wide variety of conventional methods have been de-veloped to remove heavy metal levels such as Iron Fe and Mn in coal mine acid drainage liquid waste, like chemical precipitation [3], coagulation/flocculation [4], use of micro-organisms, adsorption [5], and passive treatment methods [6]. Of all these methods, the treatment method widely used for acid mine drainage is the passive treatment method, which uses a constructed wetland system [7]. In the wetland constructed system, three processes can occur that occur nat-urally, namely chemical, physical, and biological processes, this is due to the interaction between the plants that make up wetlands and the environment which contributes to the in-crease in organic matter content through substances result-ing from the secretion and decomposition of plant residues that can help stabilize the substrate, and maintain microbial populations [8].

The system constructed wetland is a system that utilizes plants as indicators of heavy metal absorbers, where in this system plants can be used as phytoremediation agents of acid mine drainage, this is because the plant has a high tolerance to metals, a root system that is resistant to pollutants, and has large biomass [9]. Some studies of aquatic plant species that have the potential to be hyperaccumulators in acid mine drainage because they can absorb contaminants are *Eichornia sp, Cyperus odoratus, Hydrilla Vercilata*, *Ipomea aquatic*, dan *Pistia Strarariotes* and *Eichhornia crassipes* [10], *Eichhornia crassipes, Eleocharis dulcis* [11][12][13][14], *Typha angustifolia* and *Fungi mikoriza arbuscular* [15].

Another plant that has the potential to be an indicator of heavy metal absorbents is the *L. articulate*. The same is the case with *Eleocharis dulcis*, a plant of *L. articulate* that also belongs to the *Cyperaceae family*, however, in different species and taxonomic sciences it is a typical plant that lives in wetlands although *L. articulate* can also live in brackish areas so that based on phytochemical screening *L. articulate* contains varied active compounds, such as active compounds of tannins, saponins, flavonoids, alkaloids, and steroids[16]. The fundamental difference between *L. articulate* and *Eleocharis dulcis* is that it is larger in shape and size and has harder leaves such as woody and more pronounced veins. In addition, *L. articulate* has a cavity on the stem that is similar to a bamboo stick, a rhizome that is brownish to blackish in color, a rougher surface, and a higher cell density [17].

From the explanation above, the study is to find out whether *L.a articulate* plants can increase the degree of acidity (pH) in coal mine acid drainage and to determine the effectiveness of *L. articulate* plants in absorbing the concentration of heavy metals Fe and Mn dissolved with passive treatment methods which in their processing use a Laboratory-scale wetland constructed system.

2. Materials and Methods

2.1 Location and Study

The study was carried out in Nursery Park PT. Bukit Asam, Tbk, Tanjung Enim, South Sumatera which will be held from May to July 2022. The location for sampling *L. articulate* plants is in the Pedamaran area, Ogan Komering Ilir districts, South Sumatera, which is at the coordinate point -3.484725” S, 104̊ 834639” E. For sampling, acid mine drainage is in the area Banko PIT 1 Utara PT. Bukit Asam, Tbk with coordinate points 3̊ 46’ 8.843” S, 103̊ 49’ 54.244” E 345̊ N.

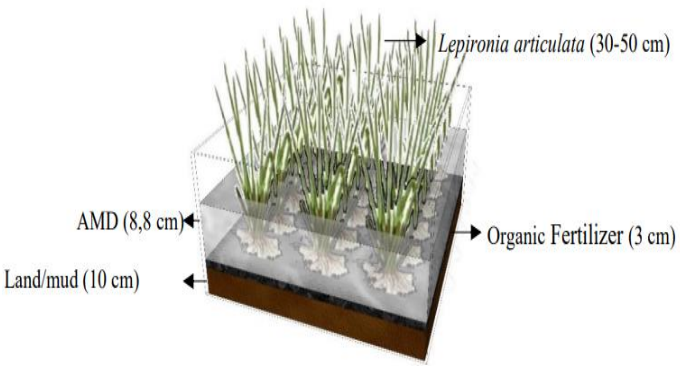
2.2 Tools and Materials

The tools used in this study were aquariums as laboratory scale wetlands with dimensions of 60 x 60 x 50 cm, mediatech digital pH meter PH-009, litmus paper, analytical balance sheet, Erlenmeyer, filter paper (Whatman), hot plate, measuring flask, 1-liter jerry can, 35-liter jerry can, large basin, ruler, scale, drip pipette, stirrer, camera, bulk, beker glass, X-ray Fluorescence Spectroscopy (PANalytical Epsilon 3 XLE XRF), oven, drum, and beaker equipment, split injection syringe 50 cc.

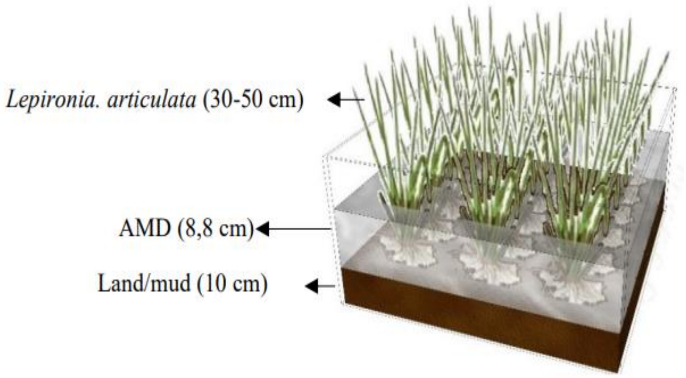
Some of the materials used in this study are *L. articulate* plants, acid mine drainage, soil or mud, organic fertilizers, equates, and water.

2.3 Research Procedure

Acid mine drainage neutralization in this study was carried out for 30 days using the passive method of treatment system constructed wetland. The type of artificial wetland pond used is an anaerobic wetland measuring 60 x 60 x 50 cm, namely reactor organic and reactor inorganic. Observation time consists of 3 stages, namely 10, 20, and 30 days with the variables measured are the degree of acidity (pH), the concentration of Fe and Mn metals in acid mine drainage, and those absorbed by plants *L. articulate*.



(a) Reactor Organic



(b) Reactor Inorganic

Figure 1. Passive Treatment Processing Trial Pool Design.

Table 1. Material Composition of Artificial Wetlands in Acid Mine Drainage.

Reactor	Treatment (Days)	Land (Kg)	Organic Fertilizer (Kg)	Plant Weight (Kg)	The Abundance of Plants
Organic	10	39.5	1.5	950 g	30
	20	39.5	1.5	950 g	32
	30	39.5	1.5	950 g	38
Inorganic	10	39.5	-	950 g	30
	20	39.5	-	950 g	35
	30	39.5	-	950 g	32

2.4. Stages of Research

The stages in this study began with sampling the Plant *L. articulate* which is used as a phytoremediation agent taken from the same growing place with a height criterion of ± 50 cm. The process of acclimatization of plants was carried out for 7 days before being used in the study, this aims to make plants able to adjust to the growing environment in phytoremediation treatment [15]. Meanwhile, for sampling acid mine drainage samples are taken directly at the Banko PIT 1 Utara PT. Bukit Asam, Tbk. The initial sample size of acid mine drainage was performed in situ and ex-situ for testing of physical and chemical properties. The parameters measured include the degree of acidity (pH), as well as the metal content of Fe and Mn.

After carrying out the acclimatization process for 7 days, the next stage is to carry out a phytoremediation process for 30 days which is carried out in 6 reactors with 2 treatments (organic and inorganic) and 1 repetition. Acid mine drainage and plant water sampling were carried out within 10, 20, and 30 days of observation[11]. The drainage acid mine sampling is carried out with randomized and Composite Samples using a 50 cc split injection syringe, then measurements of acidity concentration (pH), Fe and Mn metal content dissolved in acid mine water, and accumulation of Fe and Mn metals found in *L. articulate* plants. For pH measurements are carried out in situ during the phytoremediation process, the measurement of the concentration of Fe and Mn metals dissolved in acid mine drainage is to use the Atomic Absorption Spectrophotometry (SSA) method, and the measurement of the composition of iron (Fe) and manganese (Mn) metals absorbed by *L. articulate* plants is carried out by the X-ray Fluorescence Spectroscopy (PANalytical Epsilon 3 XLE XRF) method exists at the UNP Laboratory (Padang State University).

Calculation of the amount of efficient removal of increase in pH and decrease in the concentration of iron (Fe) and manganese (Mn) metals in coal mine acid drainage using equations 1 and 2 [18]. Then the data is analyzed to make conclusions based on the research objectives.

$$E = \frac{C_0 - C_e}{C_0} \times 100\% \qquad (1)$$

Where:

C_0 = pH optimal
C_e = initial pH
E = Percent increase in pH (%)

$$E = \frac{C_0 - Ce}{C_0} \times 100\% \qquad (2)$$

Where:

C_0 = Initial levels of Metals Fe and Mn (mg/L)
C_e = The final content of metal iron (Fe) and manganese (Mn) (mg/L)
E = Percent decrease (%)

3. Results and Discussion

3.1. Reduction in the degree of acidity (pH) by the plant *L. articulate*

The degree of acidity (pH) is an indicator to find out the level of acidity or wetness of a solution and its changes can affect existing biota or microorganisms [19]. The initial pH value of drainage acid mine in the mine sump is 2.8. However, after the testing process using wetland construction, it was recorded that the pH value increased from the initial pH, as presented in Figure 1. This shows the effectiveness of the plant *L. articulate* can increase the degree of acidity (pH).

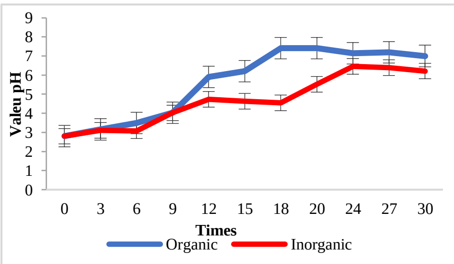


Figure 2. increasing the pH of Organic and Inorganic Reactor Mine Acid Drainage

The results of observations for 30 days in each reactor experienced a gradual and dynamic increase in acidity (pH) with values of 3.15 – 7.41 in organic reactors and 3.08 – 6.46 inorganic reactors. These results show that the pH of each reactor is following the quality standards, namely 6-9. The difference in values obtained from each treatment is clearly evident, where the organic fertilizer reactor is higher compared to the reactor without organic fertilizer. This occurs because of the presence of organic materials such as proteins, carbohydrates, and fats in organic fertilizer that can accelerate growth and increase the activity of sulfate-reducing bacteria that potentially affect the pH of acid mine water in phytoremediation processes. [7].

As with *Eleocharis dulcis*, *L. articulate* also contains alkali and alkaline earth metals such as K and Ca which if reacting with water will form alkaline compounds[20]. The reaction of the formation of alkaline compounds can be seen in the following equation.

$$2K^{+}_{(aq)} + H_2O_{(l)} \longrightarrow 2KOH\ (aq) + H_2\ (g) \quad (1)$$

$$Ca^{2+}_{(aq)} + 2H_2O_{(l)} \longrightarrow Ca\ (OH)_2\ (aq) + H_{2\,(g)} \quad (2)$$

From the equation of the compound above, which is alkaline if it reacts with H_2SO_4, it will form a neutralization reaction like the following equation.

$$2KOH_{(aq)} + H_2SO_{4\,(l)} \longrightarrow K_2SO_{4\,(aq)} + 2H_2O_{(l)} \quad (3)$$

$$Ca\ (OH)_{2\,(aq)} + H_2SO_{4\,(l)} \longrightarrow CaSO_{4\,(aq)} + 2H_2O \quad (4)$$

The results of the percentage calculation for the increase in optimum pH in the reactor organic were 62.21% and 60.94% reactor inorganic. The high percentage of reactor organic is due to the addition of planting media that contains alkali and alkaline earth metals such as K and Ca. If alkali and alkaline earth metals react with water, it will form an alkaline reaction. This is because of an increase in the pH of acid mine drainage in organic reactors higher than that of inorganic reactors. The compound of reaction formation can be seen in the equation.

$$2\ K^{+}_{(aq)} + 2\ H_2O_{(l)} \longrightarrow 2\ KOH_{(aq)} + H_{2\,(g)} \quad (5)$$

$$Ca^{2+}_{(aq)} + 2\ H^2O_{(l)} \longrightarrow Ca\ (OH)_{2\,(aq)} + H_{2\,(g)} \quad (6)$$

From the equation of the compound above which is alkaline if it reacts with H_2SO_4 will form a neutralization reaction like the following equation.

$$2\ KOH_{(aq)} + H_2SO_{4\,(aq)} \longrightarrow K_2SO_{4\,(aq)} + 2\ H_2O_{(l} \quad (7)$$

$$Ca\ (OH)_{2(aq)} + H_2SO_{4\,(aq)} \longrightarrow CaSO_{4(aq)} + 2\ H_2O \quad (8)$$

3.2. Effect of *Lepironia articulate* on Reducing Iron (Fe) and Manganese (Mn) Metal Levels in Coal Mine Acid Drainage

Regulation of the Ministry of Environment No. 5 of 2023 and Decision of the Governor of South Sumatra No. 8 of 2012, about Water Quality Management and Water Pollution Control, requires iron metal (Fe) content to be 7 mg / L and Manganese (Mn) 4 mg/L as a parameter of wastewater derived from mining activities.

In this study, the phytoremediation process was carried out using two variations of treatment, namely using organic fertilizer as a matrix enhancer and without organic fertilizer, as well as temporal variations in sampling so that they can make the most of the ability of plants to absorb dissolved heavy metals in coal mine acid water waste. Plants have different levels of sensitivity and abilities in eliminating heavy metals. The ability of the *L. articulate* plant to accumulate heavy metals can be seen from the decrease in the concentration of heavy metals Fe and Mn dissolved in water, where plants carry out phytoremediation mechanisms by *rhizofiltration* and *phytoextraction*, namely plant roots attract metals, accumulate, precipitate and translocate all parts of the plant.

Removal Efficiency of Heavy Metal Iron (Fe) Acid Mine Drainage

Analysis of Fe metal content in coal mine acid water aims to determine the decrease in concentration and removal efficiency by *L. articulate* in each reactor. Sample measurements were carried out every 10 days for 30 days. From the results of the analysis that has been carried out, *L. articulate* can remediate water polluted with Fe metal. This can be seen from the amount of optimum value produced. In organic reactors, *L. articulate* can absorb the maximum concentration of Fe metal on day 20, which is <0.006 mg/L and 0.04 mg/L inorganic reactor. However, the increase occurred on the 30th day in each of the reactors. This is due to high dissolved Fe^{2+} at low water pH [21]. Changes in Fe concentration can be caused by several factors, such as changes in water pH, sulfide minerals oxidized from sediment to water, and organic matter [22].

The oxidation reaction of the mineral sulfide (*pyrite*) can be seen in the following equation.

$$2\ FeS_2 + 7\ O_2 + 2\ H_2O \rightarrow 2\ Fe^{2+} + 4\ SO_4^{2-} + 4\ H^+ \quad (9)$$

$$FeS_2 + 14\ Fe^{3+} + 8\ H_2O \rightarrow 15\ Fe^{2+} + 2\ SO_4^{2-} + 16\ H^+ \quad (10)$$

$$4\ Fe^{2+} + O_2 + 4\ H^+ \rightarrow 4Fe^{3+} + 2H_2O \quad (11)$$

$$4\ Fe^{2+} + O_2 + 10\ H_2O \rightarrow 4\ Fe\ (OH)_3 + 8\ H^+ \quad (12)$$

$$4\ FeS_2 + 15\ O_2 + 14\ H_2O \rightarrow 4\ Fe\ (OH)_3 + 8\ SO_4^{-2} \quad (13)$$

This reaction is a source of increase or decrease in ferrous ions to release acid into the environment. Sulfate ions and the acidity formed can affect the acidity level of water. Because ferrous iron ions as strong oxidizers can dissolve sulfide minerals the metals contained in sulfide minerals are dissolved.

Table 2. Iron (Fe) Metal Removal Efficiency

Reactor	Initial Sample	Day to-	Fe Heavy Metal Removal Efficiency (%)	
			Impairment Value (mg/L)	Decreased Efficiency (%)
Organic	0.09	10	0.018	80
	0.09	20	<0.006	93.33
	0.09	30	0.013	85.56
Inorganic	0.09	10	0.058	35.56
	0.09	20	0.039	56.67
	0.09	30	0.040	55.56

Table 2, showed the optimum value of the decrease in the concentration of Fe metal that occurred on the 20th day, which was <0.006 mg / L with a removal efficiency of 93.33% in organic reactors and 0.039 mg / L with a removal efficiency of 55.67% for inorganic reactors. *L. articulate* plants can absorb Fe metal to the maximum on the 20th day and on the 30th day there is a decrease in the removal efficiency value in each reactor, this is due to the large amount of dead or decaying plants, where their organic matter will be decomposed by microbes present in the water. This decomposition process can produce organic compounds that can release Fe ions into the water. In addition, soil particles can re-adsorb the dissolved Fe metal. When plants absorb Fe metal, a chemical balance shift occurs in the soil, causing Fe metal that is bound to soil particles to become dissolved in water. This can cause the concentration of Fe in the water to increase again after a decrease due to the phytoremediation process. [23].

The decrease in the value of removal efficiency occurs along with the increase in exposure day and the ability of plants to absorb heavy metals and utilize them for growth [24]. But, several other possibilities can cause a decrease in the efficiency value of Fe removal in acid mine drainages, such as the interaction between Sulfide (S^{2-}) produced by the sulfate reduction process with metals with 2 valence metals (Fe^{2+} and Mn^{2+}), the existence of a metal adsorption process by organic matter, and plant tissues, as well as the process of metal biosorption by microorganisms found in a wetland environment[7].

Efficiency Removal of Manganese (Mn) Heavy Metal Acid Mine Drainage.

The analysis of the decrease in manganese (Mn) levels in acid mine drainage was carried out the same as the treatment of iron metal Fe which aimed to determine the value of the decreasing concentration and efficiency removal of dissolved metal Mn levels absorbed by the plants. Measurements are carried out every ten days to 30 days. The measurement results can be seen in Table 3.

Table 3. Manganese (Mn) Metal Removal Efficiency

Reactor	Initial Sample	Day to-	Heavy Metal Removal Efficiency Mn (%)	
			Impairment Value (mg/L)	Decreased Efficiency (%)
Organic	8.1	10	7,56	6,67
	8.1	20	5,02	38,02
	8.1	30	7,86	2,46
Inorganic	8.1	10	7,95	1,85
	8.1	20	6,01	25,8
	8.1	30	6,32	21,97

From the results of measuring the metal content of Mn acid mine drainage, the decrease in Mn metal content occurred in each treatment in each reactor, although not so significant. Table 4.3 shows the ability of *L. articulate* plants to absorb Mn metal, the highest rate of decrease in Mn metal concentration occurred on day 20 at 5.02 mg/L with a removal efficiency of 38.02% organic reactor and 6.01 mg/L with a removal efficiency of 25.8% inorganic reactor.

This is because of several factors, such as the influence of plants in absorbing Mn metal and the addition of media (organic substrate) which plays an important role in reducing metal concentration, where the addition of organic substrate provides a surface for bacterial activity and also helps plant growth.[17]. The addition of organic substrate as a matrix to the organic reactor plays an important role in reducing metal concentration. Adding organic substrates provides a surface for bacterial activity, helps plant growth, and can release electrons to bind atoms due to the reactive composition of organic substrates stimulating sulfate predictive materials that can increase alkalinity and convert metals in the form of disulfide deposits, and become an energy source for sulfate predictive bacteria.[25].

The increase in concentration and removal efficiency of Fe and Mn metals occurred on day 30 in both organic fertilizer reactors and non-fertilizer reactors. This condition happened because the plants had reached saturation point and were no longer able to absorb Fe and Mn metals to the maximum. However, even though the plants were in a saturated state, they would not release heavy metals that had been absorbed in the form of dissolved metals. Plants can adapt by shedding metals that are attached to the roots, and the released metals will settle together with the substrate in a simpler form and not in the form of dissolved metals as before. [26].

4. Conclusion

The conclusion obtained in this study is that the *L. articulate* plant can increase the degree of acidity (pH) and reduce the concentration of ferrous metals (Fe) and manganese (Mn) dissolved in acid mine water, this can be seen from the increased in the initial pH of 2.8 to 7.41 in organic reactors and 6.46 inorganic reactors, with a removal efficiency of 62.21% and 60.94%. The increase in pH is influenced by the content of Ca and K in *L. articulate* that if it reacts with water, it will form alkaline compounds. The decrease in metal levels of Fe and Mn occurred on the 20th day of the experiment. Reduction in Fe metal content of <0.006 mg/L with a removal efficiency of 93.33% in organic reactors and 0.039 mg/L with a removal efficiency of 55.67% for inorganic reactors, and a decrease in Mn levels of 5.02 mg/L with a removal efficiency of 38.02% organic reactors and 6.01 mg/L with a removal efficiency of 25.8% inorganic reactors.

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