



Fe and Mn phytoremediation of acid coal mine drainage using water hyacinth (*Eichornia crassipes*) and chinese water chestnut (*Eleocharis dulcis*) on the constructed wetland system

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Abstract

Acid Mine Drainage (AMD) is a wastewater formed through a series of chemical reactions and biological activities during and after open-pit system coal exploitation. Coal containing sulfide in the presence of oxygen and air is oxidized to form sulfuric acid, thus having a pH < 4. This condition facilitates the solubility of Fe and Mn. As a result, AMD has a great potential as environmental pollutants. This study aims to determine the efficiency of Fe and Mn removal on AMD and potency of water hyacinth and Chinese water chestnut to accumulate Fe and Mn. The method used is phytoremediation by water hyacinth/ eceng gondok (*Eichornia crassipes*) and chinese water chestnut/ puruntikus (*Eleocharis dulcis*) on constructed wetland system (CW). The treatment was carried out for 25 days with a flow rate of 5 m³/day. Measurements and samplings are done every 5 days. Measurements of Fe and Mn concentrations using ICP-OES. The results show that the CW is only able to increase the pH from 3.20 to 5.31. Water hyacinth and chinese water chestnut are able to accumulate Fe and Mn with the highest Bioconcentration Factor (BCF) for Fe, respectively from 1701.12 and 1010.86 and for Mn, respectively 1.12 and 1.45. Phytoremediation Index (PRI) or the CW performance efficiency in Fe and Mn removal respectively between (87.11– 95.28) % and (70.08 – 79.84) %. These results indicate that both plants can be considered to be utilized for long-term AMD processing in wider CWs.

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Introduction

Acid Mine Drainage (AMD) is a wastewater formed through a series of chemical reactions and biological activities. Rocks containing sulfides in the presence of oxygen and water are oxidized to form sulfuric acid, thus having a pH <4. At low pH, the heavy metal solubility in water is increased. As a result, AMD with high acidity and rich in heavy metals has great potential to be a source of environmental pollution. Some researchers have found that AMD has a low pH, high sulfates, and contains toxic heavy metals such as Fe, Mn, Pb, Cd, Hg, As, Al, Cr, Ni, Zn, Co, Cu (Achterberg *et al.*, 2003; Dowling *et al.*, 2004; Blodau, 2006; Elisa *et al.*, 2006; Yunus, 2014; Prihatini *et al.*, 2015; Prihatini *et al.*, 2016a; Prihatini *et al.*, 2016b; Prihatini and Soemarno, 2017).

Chemical methods that can be used to remove excessive heavy metals from water are coagulation, ion exchange, precipitation, electrolysis, and reverse osmosis system (Balasubramanian *et al.*, 2009; Kim *et al.*, 2006; Kumari *et al.*, 2006). Currently, these methods are rarely used because of having several weaknesses such as to require knowledge, skills and high cost (Sharma and Sohn, 2009). In the South Kalimantan mining industry nowadays, the method used is by adding limestone to achieve a government-set quality standard (pH 6-9), TSS (200), total Fe (7 mg/L max.), total Mn (4 mg/L max.), and Cd (0.05 mg/L max.) (Environment Ministerial Decree No: 113 for 2003). This method is also considered expensive and leads to high deposition of inorganic sludge.

South Kalimantan is largely a marshland with diverse water plant vegetation. The most dominant vegetation is Chinese water chestnut (*Eleocharis dulcis*) and Water hyacinth (*Eichornia crassipes*). Both water plants can grow and develop in swamp area and classified as heavy metal hyperaccumulator. Krisdianto *et al.* (2006), found that chinese water chestnut could function to decrease the concentration of dissolved Fe in rice planted plots using coal AMD sources, yielding Fe uptake with an average of 1176.6 mg/kg dry weight. Prihatini *et al.* (2015), found that chinese water chestnut in horizontal subsurface flow

constructed wetland (CW) system which operated by continuous flow had AMDFe removal efficiency by 78.05%.

Similar thing with water hyacinth, this plant able to accumulate several metals, such as Al, Pb, Cu, Fe, Mn, Ni, Cd, Cr, Co, Zn, and Hg (Skinner *et al.*, 2007). Water hyacinth can accumulate soluble As at AMD (Michelle *et al.*, 2010). Recent studies have shown that water hyacinth is able to reduce Pb and As simultaneously at AMD with phytoremediation index (PRI) respectively by 100% and 70.51% of the initial concentration of 10.20 ppm and 12.34 ppm respectively and can increase pH from 4.87 to 6.85 (Yunus, 2014). The data on both plants capabilities are considered for plant use as heavy metal accumulators and to increase the pH of AMD in CW construction.

The CW construction has been used internationally with satisfactory results (Farooqi *et al.*, 2008). In 2000 alone, there were 600 active constructed wetland projects in the United States (USEPA, 2000) and more than 400 active projects in Europe. In Indonesia, the CW has been used to improve the water quality of the Citarum River (Meutia *et al.*, 2003). In addition, a study was conducted at the Surabaya Urban Community Empowerment Center in 2007 to see the CW's ability to treat domestic wastewater. This study demonstrates that the built CW has relatively lower capital and operating costs than conventional waste processing with equivalent performance, making it an attractive alternative (Soewondo and Akbar, 2007). The studies also indicate that CW can be applied to the mining industry in processing other types of waste including coal AMD. Therefore, in this study we want to know the effectiveness of constructed wetland system to treat coal AMD and potency of water hyacinth also Chinese water chestnut as phytoremediation plants in constructed wetland.

Materials and methods

The materials used were obtained from the vicinity of the coalmine site. Water hyacinth (Wh) that were used

have heightsof 20-25 cm with perfect condition, chinese water chestnut (Cwc) tillers that have heights of about 15-20 cm, and manure.

Continuous flow CW was made to the size of 2 × 30 m or with an area about 60 m² (Fig. 1a). CW was filled with soil mix with 10% bokashi (Fig. 2a). This CW was made into two parts, of which half was planted with chinese water chestnut and the other half was planted with water hyacinth. Both planted plants have a relatively same number of stems per clump with a distance of about 15 cm (Figure 2b). Plant acclimatization was done by providing clean water for 2 weeks or until vegetation cover >70% with the mortality rate <10%. Continuous CW was operated for 25 days with a flow rate of 5 m³/day (Figure 4).

AMD to be processed was sourced from void of one of the coal mining companies in South Kalimantan (X Inc.)

Sampling for Fe and Mn measurements contained in (water, water hyacinth, and chinese water chestnut) were done every 5 days from day 0-25. Water samples were taken at 2 points, each in inlet and outlet of 100 mL with 3 replications. Samples from both plants were also taken at 2 points near the inlet and near the outlet. Each sampling was preceded by pH and water temperature measurements.

Sample analysis (water, Wh, and Cwc) on Fe and Mn content was performed in accordance with SNI standard, that is SNI 6989.4.2009 for Fe and SNI 6989.5.2009 for Mn. Measurements of Fe and Mn concentrations were done using Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) ACTIVA S.

Table 1. AMD quality.

Parameters	Unit	Analysis result	Maximum level Environment Ministerial Decree 113/2003
Fe concentration	ppm	23.12	7
Mn concentration	ppm	25.50	4
pH	-	3.20	6-9
TSS	ppm	37.05	400

Data analysis

The measurement results of Fe and Mn concentration accumulated on Wh and Cwc were converted to Bio-concentration Factor (BCF).

BCF is a parameter to determine the potential of plants as Fe and Mn accumulators in dry weight conditions of plants. BCF is calculated by the formula (Zayed *et al.*, 1998):

$$BCF = \frac{\text{The concentration of metals in plants}}{\text{Metal concentration in AMD}}$$

The results of measurements of Fe and Mn concentrations contained in the AMD were converted to the value of the Phytoremediation Index (PRI).

PRI is the decrease of percentage in initial parameter concentration compared with parameters on effluent. PRI is calculated by the following formula:

$$PRI = \frac{[\text{initial}] - [\text{final}]}{[\text{initial}]} \times 100\%$$

Results and discussion

Acid mine drainage quality

Table 1 shows that based on the Environment Ministerial Decree No. 113/2003 and South Kalimantan Gubernatorial Regulation No. 36/2008 on quality standard requirements of coal mine wastewater; there are three unqualified parameters, ie Fe, Mn, and pH concentrations. Very high concentrations of Fe and Mn with very low pH indicate that the AMD should be treated before being released into the environment.

TSS parameter is already up to the standard so that it's no longer analyzed.

Water hyacinth and chinese water chestnut bio-accumulation

Table 2 shows that Wh and Cwc are able to absorb and accumulate Fe and Mn from AMD at very low fluctuating temperatures. The accumulation of both plants also appears to be incompatible with the

increase in pH. Changes in pH from 3.20 to 5.31 or increased by 2.1 over a 25-day time span in a continuous flow CW system show that the pH of AMD has not met the standard of coal wastewater determined by the Environment Ministerial Decree No 113/2003 of 6 -9.

Table 2. Measurements of Fe and Mn Concentrations Accumulated in Water Hyacinth and Chinese water chestnut (mg/kg dry weight).

Parameters	Treatment time (Day)					
	0	5	10	15	20	25
Temperature (°C)	37.2	37.3	37.5	37.2	37.1	37.7
water outlet pH	3.20	3.97	4.64	4.70	4.96	5.31
Fe conc.onWh	1946.79	10263.00	5468.77	39329.83	7734.46	5986.11
Fe conc.onCwc	3709.87	23371.00	20071.08	18858.59	9229.45	19935.26
Mnconc.onWh	4.03	10.28	7.95	23.89	26.93	28.56
Mnconc.onCwc	5.88	27.43	7.93	3.18	36.86	29.06

The increase in pH has proven that AMD treatment with CW utilizing Wh and Cwc may increase the AMD pH. This increase in pH can be caused by the interaction of settling process, sedimentation,

adsorption, co-precipitation, cation exchange, photodegradation, phytoaccumulation, biodegradation, microbial activity, and plant uptakes (Sheoran and Sheoran, 2006).

Table 3. Processed water products quality at CW.

Parameters	Treatment time (Day)						Average
	0	5	10	15	20	25	
pH	3.20	3.97	4.64	4.70	4.96	5.31	4.72
Fe concentration(ppm)	23.12	1.67	1.66	1.09	2.98	1.31	4.15
MNconcentration(ppm)	25.50	7.48	5.14	7.66	5.36	7.63	6.65
Fe PRI (%)		92.78	92.82	95.29	87.11	94.33	92.47
MnPRI (%)		70.67	79.84	69.96	78.98	70.08	73.91

The accumulation of Fe and Mn from Wh and Cwc on the CW system shows an unstable increase. Water hyacinth could accumulate the highest Fe and Mn of 39329.83 mg/kg dry weight on day 15 and 28.56 mg/kg dry weight on day 25 respectively. Chinese water chestnut were able to accumulate the highest Fe and Mn respectively, of 23371.00 mg/kg dry weight on day 5 and 36.86 mg/kg dry weight on day 20. The trend of Fe and Mn accumulation rate from both plants shown in Fig. 1 and 2.

BCF calculations showing the potential of both plants in accumulating Fe and Mn are shown in Figs. 3 and 4. Fig. 3 shows that Wh has Fe BCF (1701.12) higher than Cwc (1010.86), whereas in the average results, Cwc (791.22) higher than Wh (595.00) occurs.

The highest Fe BCF each occurred on the 15th day for Wh and the 5th day for Cwc.

The high Fe BCF value indicates that Wh and Cwc deserve to be called the Fe hyperaccumulator. The same phenomenon has also been demonstrated by previous studies (Prihatini and Soemarno, 2017).



Fig. 1. Sampling locations (a) Water hyacinth, (b) Chinese water chestnut.

This result explains that Cwc has an adaptation time to environmental conditions (pH) shorter than water hyacinth in adsorption Fe from AMD but not on transport and translocation process. This is apparent after the highest BCF occurs; Cwc takes longer to improve the ability of its adsorption.

Based on Fig. 4, the Mn BCF of both plants is much lower than that of Fe BCF. The highest Mn BCF of Wh (1.12) and Cwc (1.45) each occurred on the 25th and 20th days. The average BCF result of both plants shows that Wh is lower than Cwc, ie 0.77 and 0.82 respectively. These results explain that both plants are not able to accumulate Fe and Mn simultaneously with high concentrations.



Fig. 2. (a) Bokashi allotment in CW, (b) CW with Wh and Cwc plants.

The tendency in Figures 3 and 4 can be explained that Cwc is a plant that can grow well in $\text{pH} < 4$, whereas Wh will experience growth restriction at $\text{pH} < 4$. This is in line with previous research that water hyacinth cannot tolerate low pH medium and best growth in the pH range of 5.5 to 7.0 (Ratnaningsih, *et al.* 2010; Lu, 2009). This causes Wh to require time and higher pH conditions to be able to perform higher adsorption. Another cause is that in $\text{pH} < 4$, dissolved cations are restricted to approach plant tissue.

According to Lopez *et al.* (2002), the binding of metals to plant tissue increases with increasing pH. This is reinforced by Kauret *et al.* (2012), that at low pH, the reacting force of the H^+ ions from the adsorbent site inhibits the metal cations. The results of the studies on different metals found that the accumulation of Zn, Pb, As, Fe and Cd by water hyacinth and chinese water chestnut increased with increasing pH (Aisen *et al.*, 2010; Yunus, 2014, and Prihatini *et al.*, 2016a, Prihatini *et al.*, 2016b,

Prihatini and Soemarno, 2017). Bioconcentration factor values (BCF) of Fe in *Eleocharis dulcis* that planted on VSSF-CW showed that *E. dulcis* is a Fe accumulator plant.

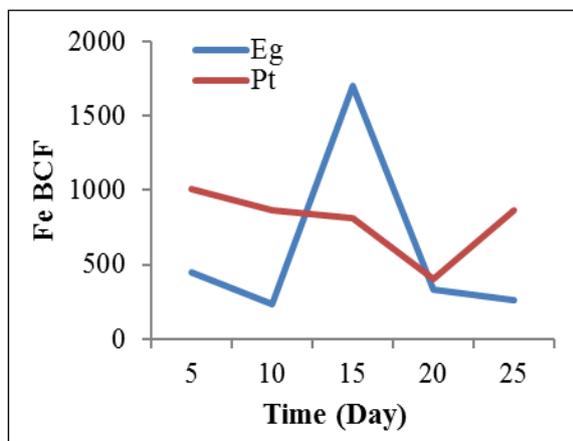


Fig. 3. Fe BCF in Water hyacinth (Wh) and Chinese water chestnut (Cwc).

Another trend is the high Fe BCF rather than Mn. This can be caused by several factors such as the electronegativity and the different metal covalent indices. Huheey *et al.* (1993), has written electronegativity based on Pauling scale, where Fe is 1.8 and Mn 1.5 with a radius of Fe ions 0.79 Å and Mn 0.75 Å. In plants, the existing groups are carboxylic and hydroxyl. The electronegativity of these two groups is 3.04 and 2.82 respectively. According to Jasmadi *et al.* (2002), that smaller ions will interact more strongly with ligands because they have high polarization capabilities. Chojnacka (2013) that the important parameters in the maximum cationbio sorption depend on the covalent index. A higher covalent index, potentially forming covalent bonds with biological ligands (thiols, amino, carboxyl, and hydroxyl). These results indicate that the high Fe BCF rather than Mn cannot be explained by the formation of the complex.

Another common theory used to explain the above phenomenon, is based on the acidity of the two metals. Pearson (1963) in Huheey *et al.* (1993), put forward the principles of HSAB (Hard and Soft Acid Bases) by classifying Lewis base acids by their strong properties and weak cations. Strongly acidic cations will interact strongly with strongly alkaline ligands. In contrast, weakly acidic cations will interact strongly

with weakly alkaline ligands. This theory also apparently cannot explain with certainty because the cation grouping according to the principle of HSAB, Fe and Mn ions are in the same group. Thus, this result is likely due to the presence of some metals present in the AMD, which have higher reactivity than Mn and which are both tolerable by both plants.

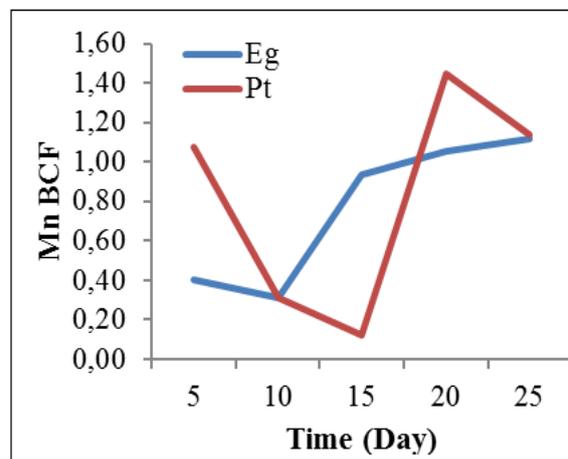


Fig. 4. Mn BCF in Water hyacinth (Wh) and Chinese water chestnut (Cwc).

CW Performance Efficiency

Table 3 shows that water quality obtained from AMD treatment with the phytoremediation method at CW within 25 days, there are parameters that have not fulfilled the requirement stipulated by Environment Ministerial Decree of No 113/2003, that is pH (<6) and Mn concentration (> 4 ppm) and only Fe concentrations have met the requirement (<7 ppm). This result also corresponds to high Fe BCF value and Mn BCF which is still very low.

The tendency to the concentrations decrease of Fe and Mn before and after through CW is shown in Table 3. The decrease in Fe and Mn concentrations appears to be non-constant. Mn concentrations on days 15 and 25 appear to increase. The same thing happens in the Fe concentration on the 20th day, which showed an increase. The increased concentration of these two metals can be caused by the accumulation of both plants on the same day decreased. Other causes by Tarutis *et al.* (1999) that CW at any given time can act as a source of metal rather than as an absorbent. This phenomenon can also be caused by AMD at low pH resulting in

increased metal desorption and mineral solubility resulting in increased concentrations of toxic heavy metals. In addition, at pH 2.3 -3.5 Fe(III) is unstable in solution and Fe(III) oxyhydroxide is insoluble and Fe(III) oxy hydroxy sulfate precipitates (Woulds and Ngwenya, 2004).

PRI values are also a measure of the CW efficiency performance. Based on the calculation of PRI showed the decrease of Fe and Mn concentrations, respectively between 87.11-95.28% and 70.08 – 79.84% or decreasing average respectively 92.47% and 72.91%. Fe and Mn PRI changes are also not constant, as shown in Table 3. This may be due to the ability of Wh and Cwc in adsorbing and accumulating Fe and Mn. Another factor is dependent on the concentration and dynamics of metals in the wastewater and the hydraulic load entering the CW (Vymazal, 2003).

Conclusion

Water hyacinth and chinese water chestnut were able to accumulate Fe with BCF respectively of 1701.12 and 1010.86 whereas both were able to accumulate Mn with BCF of 1.12 and 1.45. CW efficiency performance in reducing Fe and Mn concentrations, respectively between (87.11-95.28)% and (70.08 – 79.84)%. The quality of water produced by the CW system using water hyacinth and chinese water chestnut within 25 days has not met the requirement of coal wastewater quality based on Environment Ministerial Decree No 113/2003. Water hyacinth and chinese water chestnut are Fe hyper-accumulator plants, so it is feasible to be recommended for use in coal AMD processing.

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