The Potential of Constructed Wetlands for Liquid Waste Management in Small and Medium-Scale Tannery: A Literature Review

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ABSTRACT

The leather tanning industry or tannery, mainly in the small and medium scale (SMEs), is not environmentally friendly. Limited capitals drive the SMEs-scale tanneries to dispose of liquid waste directly into water bodies without proper treatment. It might cause serious environmental problems due to the high content of COD, BOD, chromium, and dyes. Treatment of liquid waste using constructed wetlands has been widely used because it is efficient, cheap, and powerful. This review discusses the latest studies in the wastewater treatment of tanneries using phytoremediation techniques and constructed wetlands and their potential applications in the SMEs tanneries.

Keywords: Liquid waste, Tannery, Phytoremediation, Constructed wetland, SMEs

1. Introduction

In general, the small and medium scale (SMEs) tannery is far from environmentally friendly. It is due to a high amount of solid and liquid waste produced. The leather tanning industry requires 50 - 150 liters of water to process every 1 ton of raw leather.

During the production process, the leather tanning industry uses many chemicals such as chromium, sulfate, sodium sulfate, lime, ammonium sulfate, sodium chloride, and sulfuric acid formaldehyde, pigments, dyes, and antifungal agents. Chromium is the most widely used tanning material in Indonesia because it is cheap, the tanning process is fast, and the leather is stable. These chemicals can increase the intensity of the toxicant produced per unit output (Khan 2001).

SMEs tannery has low financial capacity. Water treatment of liquid waste requires a relatively expensive cost and might reduce the profits. Purba et al. (2020) found that the SME tanneries disposed of the liquid waste directly into water bodies without prior treatment. This practice can cause serious environmental problems because the wastewater contains high COD levels, BOD, chromium, and dyes (Song et al. 2000). The tannery's wastewater contains about 500-1000 ppm of chromium (Aravindhan et al. 2004). Chromium can poison animals and plants (Chidambaran et al. 2009).

The processing costs for the liquid waste of tannery are expensive. It has led many developing countries to use primary and (or) secondary processing in biological and physicochemical processes such as ion exchange, reverse osmosis, electrolysis systems, precipitation, coagulation, and adsorption (Kacaoba et al. 2002, Hafez et al. 2002). However, this management system is usually expensive and produces secondary pollutants. It is not economically practical for SME tanneries. According to data from the Indonesian Tanned Leather Association, 75% of the leather tanning industry in Indonesia is a small and medium scale industry. Therefore, a practical and cheaper liquid waste management technology is needed.

Processing liquid waste using constructed wetlands is widely practiced because it is efficient, cheap, and powerful (Lu et al., 2016). Constructed wetland (CW) systems have been widely used to treat various forms of liquid waste, such as industrial waste, rural household waste, urban household waste, and non-point-source pollutants (Matamoros and Salvado 2012; Galanopoulos et al. 2013; Shao et al. 2013). CW's water plants are an essential element that plays a role in treating liquid waste together with the living organisms (bacteria, fungi, yeast, and algae) in the water and fillers, forming a unique flora and fauna environment. Waste purification can occur through filtration, adsorption, sedimentation, ion exchange, absorption by plants, and microbial decomposition (Lu et al. 2016). The utilization of plants in the decomposition and purification of waste is called phytoremediation. This paper aims to review the potential for processing liquid waste from the leather tanning industry using phytoremediation techniques and constructed wetlands.

2. Discussion

Liquid Waste Characteristics of Leather Tanning Industry

Liquid waste from the leather tanning industry is generally cloudy and smelly because it contains residue of meat, blood, lime pulp, fine hairs, dissolved protein, residual salt, acid, paint residue, and chrome tanning (Yazid et al. 2007). The type of tanning material affects the levels of pollutants in the liquid waste of the tannery. Chromium is the most widely used tanning material. It will cause heavy metal residues in the liquid waste. Chromium (Cr) is a metal with oxidation values ranging from 2+ to 6+ but usually exists as trivalent chromium Cr (III) and hexavalent Cr (VI) chromium (Cheung et al. 2007). Chromium in the form of Cr(VI) has the highest toxicity level, about 10 to 100 times that of Cr (III) (Chauhan et al., 2015). The United States Environmental Protection Agency has classified Cr6+ as one of 17 chemicals toxic to humans. It is considered one of the top 20 contaminants that need to be treated before being released into the environment. The characteristics of liquid waste from the leather tanning industry from several studies and the Quality Standards for Liquid Waste for Leather Tannery Industry Activities from the Minister of Environment of the Republic of Indonesia (KEP-51/MENLH/10/1995) are presented in Table 1.

Table 1. Chemical content of tannery wastewater

Parameter	Cristina *et al*. (2007)	Singgih *et al*. (2008)	Terfie *et al*. (2015)	KEP-51/MENLH/10/1995
pH	6.14 ± 1.10	-	8.2 ± 2.3	6.0 - 9.0
BOD_5*	1000 ± 88	20	1054 ± 448	150
COD*	2250 ± 565	54	4434 ± 1846	300
TSS*	92 ± 36	20		150
Cr total*	0.027 ± 0.075	< 0.0072	40 ± 27	2.0
Cr(VI)*	0.004 ± 0.006	-	-	-
NH_3*	100 ± 14	0.58		-
NH_4^+*	-	-	563 ± 4	-

* in mg/L

Phytoremediation

Plants can reduce the number of toxic metals from the soil with microorganisms' help through assimilation and biotransformation mechanisms. For example, Cr (VI) levels in effluent can be reduced through biotransformation by *Bacillus coagulans*. Bacillus coagulans uses soluble enzymes and malate as external electron donors (Philip et al., 1998). The vascular plant absorbs toxins from the air through leaves or from soil and water through the roots. Various plant species have been identified and tested their ability to absorb and accumulate various types of toxic metals, phenolic compounds, azo dyes, various other organic and inorganic contaminants such as Cr (VI) in tannery effluent. These plant species act as excluders, accumulators, and hyper-accumulators. Excluders accumulate pollutants from the substrate into the roots and limit their movement to other parts, such as shoots.

Accumulators accumulate and convert pollutants to inert forms in aerial networks. Hyperaccumulators accumulate very high amounts of pollutants compared to other types of plants (Memon *et al*. 2001; Sinha *et al.* 2007; Memon and Schroder 2009; Sheoran *et al.* 2011; Malik and Biswas 2012). Some of the characteristics of phytoremediation as an environmentally friendly

technology are presented in Table 2. Research on the mechanism of metal absorption in plants has been widely studied (Lone et al., 2008; Sureshvarr *et al.*, 2010; Ali *et al.*, 2013).

Table 2. Phytoremediation characteristics

Advantage	Disadvantage
Autotrophic system	Low biomass and slow growth, especially types of hyperaccumulators
Requires small nutritional input, does not require a particular dump.	Longer time
Cost-effective	Limited bioavailability of pollutants
A sustainable and environmentally friendly strategy	Contamination risk on the food chain

Source: Khandare *et al.* (2011a,b); Telke *et al.* (2011;) Ali *et al.* (2013)

Phytoremediation is an inexpensive, efficient, and environmentally friendly process. This technique efficiently utilizes plants, enzymes, and microbes to isolate, transport, absorb, detoxify, and remove toxic minerals through complex biological, physiological, and chemical processes (Sureshvarr *et al.* 2010; Khandare *et al.* 2011a,b; Etim 2012; Ali *et al.* 2013). These crops are then harvested, processed, and safely discharged. This method is suitable for removing low to moderate levels of contaminants (Ghosh and Singh 2005). Based on the detoxification process, pollutant type, medium, and pollutant content, phytoremediation can be classified into phytoextraction, phytofiltration, phytostabilization, rhizodegradation, and phytovolatilization (Table 3) (Raskin and Ensley 2000; Sureshvarr *et al.* 2010; Ali *et al.* 2013). The effectiveness of phytoremediation and pollutants absorption (both organic and inorganic) is strongly influenced by plant species and their characteristics, interactions in the root zone, the nature of the medium, chemical properties of contaminants, bioavailability of contaminants, the effect of supplementing chelating agents, and environmental conditions (Cunningham and Ow 1996; Tangahu *et al.* 2011).

Constructed Wetland

Constructed wetlands (CW) are systems designed and built to take advantage of natural processes that involve vegetation in swamps, soil, and interconnected microbial assemblages to treat wastewater. CW is designed to take advantage of the processes that occur in natural swamps but a more controlled environment. CW is classified based on the most dominant aquatic plants (macrophyte) and water flow pattern.

Metal removal mechanism

Heavy metal removal mechanism occurs in the three main compartments of CW, namely (1) soil and substrate, (2) hydrology, and (3) vegetation. In general, water is stagnant on the surface or in the roots for a long time. The hydrological factor is thought to be the primary determinant of specific CW species' formation and maintenance processes. Saturation that occurs permanently or periodically will produce anaerobic conditions in the soil that cause biogeochemical processes. This process causes the development of soil characteristics in the wetland to support dominant plants' adaptation in saturated soils (Mitsch and Gosselink 1993; ITRC 2003). The hydrological compartment also contains heterogeneous polyligands, namely fulvic, humic and tannic acids, amorphous metal oxyhydroxides, clays, surface bacteria, and associated exocopolymers, suspended particles, and macromolecules, e.g., polysaccharides and proteins (Matagi *et al.* 1998).

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Table 3. Various phytoremediation processes and mechanisms

Phytoremediation mechanism	Mechanism	The procedure of contaminants absorption	Mechanism	Application	Reference
Phytoextraction	Absorbs contaminants through the roots and concentrates in the harvestable parts of the plant. For example, shoots	Through plant roots	Absorption	Inorganic pollutants, e.g., metal	McCutcheon and Schnoor (2003); Marmiroli *et al.* (2006); Ali *et al.* (2013)
Phytofiltration	Utilizes the root surface area to absorb pollutants from water	Through plant roots (rhizofiltration), young plant seeds (blastofiltration), or cut plant shoots	Filtration, absorption, precipitation of pollutants around the root zone	Inorganic pollutants, e.g., metal	Marmiroli *et al.* (2006); Macek *et al.* (2009); Al-Baldawi *et al.* (2013); Ali *et al.*(2013)
Phytostabilization	Immobilization of pollutants and their bioavailability	Through plant roots	Absorption, precipitation, and complexation in the rhizosphere	Inorganic pollutants, e.g., metal	Marmiroli *et al.* (2006); Macek *et al.* (2009); Ali *et al.*(2013)
Phytotransformation	Break down organic components through metabolic activity and plant enzymes	Through plant roots or the metabolic process around the roots	Absorption by the root system causes changes through metabolic processes or enzymes	Organic or xenobiotic pollutants	Marmiroli *et al.* (2006); Macek *et al.* (2009); Al-Baldawi *et al.* (2013); Ali *et al.* (2013)
Rhizodegradation	Degradation of pollutants through the symbiosis of microbes found in plant roots	Changes in the roots area	Secretion of exudate from roots or enzymes around the root zone and degradation by xenobiotic microbes	Organic or xenobiotic pollutants	Al-Baldawi *et al.* (2013)
Phytovolatilization	Converting pollutants and their derivative products into volatile materials or gases and releasing them into the air through transpiration	Absorption of water-soluble pollutants through plant roots	Modification of pollutants during the transport process from roots to leaves in plant vessels.	Organic or xenobiotic pollutants	Marmiroli *et al.* (2006); Macek *et al.* (2009); Ali *et al.*(2013)

The metal removal process in CW occurs through a highly complex mechanism. This process is influenced by the composition of the substrate, the sediment's pH, the characteristics of the flowing wastewater, and the plant type. The metal removal process in CW can occur through three mechanisms, namely physical, chemical and biological.

Metal removal through physical processes

Removal through physical processes can occur due to the settling and sedimentation processes. Sedimentation has long been recognized as the fundamental principle in removing heavy metals from wastewater at CW. Sedimentation occurs after another physical process, which results in the formation of heavy metal aggregates to form large particles and sink (Walker and Hurl 2002). This process starts with chemical processes such as precipitation and co-precipitation. The sediment formation rate at CW can be increased by increasing the wastewater pH, solute concentration, ionic strength, and algae concentration (Matagi *et al.* 1998). Heavy metals will be separated from wastewater and trapped in CW sediment, thus protecting the aquatic ecosystem above it.

Metal removal through a chemical process

Chemical removal can occur through sorption, adsorption, oxidation, hydrolysis of metals, precipitation and co-precipitation, metal carbonates, and metal sulfites. Sorption is the transfer of ions from water to the soil, for example, from the liquid phase to the solid phase. Sorption describes a group of processes that involve adsorption and precipitation reactions (Sheoran and Sheoran 2005).

In the sediment, heavy metals are adsorbed on soil particles due to cation exchange or chemisorption. Cation exchange occurs when cations adhere to the clay and organic materials' surface due to electrostatic attractions. When the heavy metal has been absorbed into the humic or colloidal clay, it will remain in the form of metal atoms and not decompose. Their characteristics can change over time due to changing sedimentary conditions (Batty *et al*. 2002; Wiebner *et al*. 2005). More than 50% of heavy metals can be readily adsorbed into particulate form in CW so that they can be removed from water by sedimentation. Iron, aluminum, and manganese can form water-insoluble compounds through hydrolysis and (or) oxidation at CW, causing various oxides, oxyhydroxides, and hydroxides (Batty *et al*. 2002; Woulds and Ngwenya 2004).

Precipitation and co-precipitation are essential adsorptive mechanisms in CW sediments. The formation of water-insoluble heavy metal deposits is one factor limiting the bioavailability of heavy metals in aquatic ecosystems. Precipitation is influenced by the solubility of the metal involved, Ksp, pH of CW, and the concentration of the metal ion and the relevant anion. When the concentration of cations and anions exceeds Ksp, precipitation occurs. Co-precipitation is also an adsorptive phenomenon in CW sediments. Heavy metals undergo co-precipitation with secondary minerals at CW. Copper, nickel, zinc, and manganese undergo co-precipitation with Fe oxide. Cobalt, iron, nickel, and zinc undergo co-precipitation with manganese oxide (Stumm dan Morgan 1981; Noller et al., 1994). CW with a suitable substrate can accelerate the growth of sulfate-reducing bacteria under anaerobic conditions. These bacteria will produce hydrogen sulfide in wastewater with high sulfate levels. Most heavy metals react with hydrogen sulfide to form metal sulfides, which are very water-insoluble (Stumm and Morgan 1981).

Metal removal through biological processes

The removal of metals by biological processes occurs due to phytoremediation processes by plants. Sharpe and Denny (1976) and Welsh and Denny (1979) reported that most of the metal uptake by plant tissues occurs through anionic absorption mechanisms in the cell walls. Metals cannot enter the plant directly. In biota, biological conversion occurs through the assimilation and metabolism of microorganisms that live on and around the macrophyte. Organic matter decomposition occurs by reducing and accumulating organic matter on the sediment's surface under permanently toxic water conditions. Hence, the surface sediment is responsible for capturing heavy metals from the influent (Matagi *et al.* 1998; Walker and Hurl 2002; Manios *et al.* 2003).

Microorganisms can also remove and store several heavy metals through metabolic processes. Sobolewski (1999) has reported the reduction of metals into immobilized form by microbial activity at CW. Metals such as chromium and uranium become immobilized when reduced through biological processes catalyzed by microorganisms (Fude *et al.* 1994). Schiffer (1989), Sinicrope et al. (1992), Nelson et al. (2002), and Adriano (2001) reported that chromium could be removed through bacterial activity at a rate of 40% to 84%.

Nitrogen Removal Mechanism

Nitrogen has a complex biogeochemical cycle with several biotic/abiotic transformations involving seven valences (+5 to -3) (Vymazal 2005). In the leather tanning industry's liquid waste, nitrogen is in the form of ammonium (NH_4^+) and ammonia (NH_3). The nitrogen transformations that commonly occur at CW are presented in Table 4. Various forms of nitrogen are involved in the chemical transformation from inorganic to organic compounds and vice versa. Some of these processes require energy (usually from organic carbon sources), and others release energy used by organisms for growth and survival. All of these transformations are necessary for the CW ecosystem to function correctly. Most of these chemical changes are controlled through the production of enzymes and catalysts by living organisms.

Table 4. Nitrogen transformation in constructed wetlands

Process	Transformation
Volatilization	Ammonia-N (aq) → ammonia-N (g)
Ammonification	Organic-N → ammonia-N
Nitrification	Ammonia-N → nitrite-N → nitrate-N
Nitrate-ammonification	Nitrate-N → ammonia-N
Denitrification	Nitrite-N → nitrite-N → N(g), N_2O
N_2 Fixation	N_2(g) → ammonia-N (organic-N)
Plant/microbial uptake (assimilation)	Ammonia-, nitrite-, nitrate-N → organic-N
Ammonia adsorption Organic nitrogen burial	Ammonia-N → N_2(g)
ANAMMOX (anaerobic ammonia oxidation)	

Source: Vymazal (2005)

Mathematical Model for Predicting Nitrogen Removal

Mathematical models can predict the amount of nitrogen breakdown over some time in CW types. This modeling is essential for designing a suitable CW. Mathematical models are unique for each type of CW. The characteristics of liquid waste, temperature, environmental conditions, type of plant, the type of rock used, and the flow rate of liquid waste can affect the mathematical model.

Mayo and Bigambo (2015) propose a mathematical model that describes the mechanism of nitrogen change in the horizontal subsurface constructed wetland for domestic waste. This mathematical model considers the dissolved biomass activity in wastewater, biofilm aggregates, and plant roots formed with variable concentrations of organic matter, ammonia, nitrates-nitrogen, and aggregates. The main nitrogen transformation processes considered are mineralization, nitrification, denitrification, plant absorption, plant rot, and sedimentation. Other functions that are thought to influence the model are temperature, pH, and dissolved oxygen. Equation 3 is one of the mathematical equations used to model the nitrification process by bacteria and biofilm. Polprasert and Agarwalla first developed this model (1994) called the Monod model.

$$r_n = \left[(\mu_n/Y_n + rb_1 + rb_2) \times \left(\frac{NH_4}{KN + NH_4 - N} \right) \times \left(\frac{DO}{KNO + Do} \right) \times C_T \times C_{pH} \times ON \right] \qquad (1)$$

μ_n is the maximum growth rate of Nitrosomonas (d^{-1}). Yn is the yield coefficient for Nitrosomonas bacteria in mg (VSS/mg). N and KN are the ammonia constants Nitrosomonas half-saturation (g/m^3). KNO is the oxygen constant of Nitrosomonas half-saturation (g/m^3). CT is temperature and is the dependent factor. C_{pH} is a Nitrosomonas growth inhibitor for pH. rb1 is the biofilm reaction rate constant for the aggregate (d^{-1}). rb2 is the biofilm reaction rate constant for plants (d^{-1}).

Potential Applications for Liquid Waste Treatment of Leather Tanning Industry

Processing industrial wastewater from the leather tanning industry using phytoremediation techniques in constructed wetlands has not been widely researched. Several researchers selected plant types for phytoremediation in CW (Table 5). *Typha domingensis* and *Borassus aethiopium* have the highest chromium removal efficiency, reaching 99%. Phragmites (reeds) species have the highest COD removal efficiency, reaching 85%, and *Phragmites australis* has the highest ammonium removal efficiency reaching 82.5%. Cristina et al. (2007) found that *T. latifolia* and *P. australis* had the best propagation adaptability to tannery wastewater. *Phragmites australis*, *Typha domingensis*, *Glyceria*

maxima can grow well in Indonesia. These plants have the highest ability to reduce COD, BOD, chromium, and NH4+/NH3 compared to other plants that have been tested on tannery wastewater. Therefore, these three plants could be utilized to process tannery wastewater in Indonesia. The horizontal sub-surface flow type CW construction is the most suitable type of CW for processing tannery wastewater. This type can tolerate input fluctuations, including if there is excess input. This ability is critical because, in general, the production capacity of the small and medium-scale leather tanning industry in Indonesia fluctuates throughout the year.

Table 5. Potential plant types for phytoremediation of tannery wastewater

Species	Removal efficiency (%)				Reference
	COD	BOD	Cr	NH_4^+ / NH_3	
Phragmites australis	80.9	77	97.7	82.5	Terfie *et al.* (2015)
C. alternifolius	64.8	67.5	98	64.8	
Typha domingensis	56.6	66.7	99	53.3	
Borassus aethiopum	58	66	99	80	
Canna indica	55	50	94	20	Cristina *et al*. (2007)
Typha latifolia	56.5	49.2	76.5	20	
Phragmites australis	57.6	48.6	29.4	17.5	
Stenotaphrum secundatum	54.6	46.5	52.9	17.5	
Iris peseudacorus	55	46.4	70.5	18.9	
Phragmite	80-85	-	-	-	Daniels (2001)
Phragmites	70	-	-	-	Daniels (1998)
Glyceria maxima dan *Phragmites*	82-85	-	-	-	

3. Conclusions

The literature study results indicate that applying the CW system for processing tannery waste is a promising approach for the secondary treatment stage. Some of the potential plants are *Phragmites australis*, *Typha domingensis*, and *Glyceria maxima*.

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