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SEMI-BATCH OPERATED CONSTRUCTED WETLANDS PLANTED WITH *PHRAGMITES AUSTRALIS* FOR TREATMENT OF DYEING WASTEWATER

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Abstract

The objective of present study is to evaluate the using of constructed wetland under semi-batch operation for the treatment of azo dye Acid Orange 7 (AO7) containing wastewater. The emergent plant selected in our study was *Phragmites australis*. Toxic signs were observed at the *Phragmites australis* after the addition of AO7 into the wetland reactors but it can adapt to the wastewater as shown in the increase of stem as the operation continue. Our result shows that the artificial aeration and the presence of *Phragmites australis* had a significant impact on the removal of organic matters, AO7, aromatic amines and NH_4-N. The COD removal efficiency in the aerated and non-aerated wetland reactors was 95 and 62%, respectively. The NH_4-N removal efficiency in the aerated wetland reactor (86%) was significantly higher than the non-aerated wetland reactor (14 %). All wetland reactors show high removal efficiency of AO7 (> 94%) but only the aerated wetland reactor perform better in the removal of aromatic amines.

Keywords: Constructed wetlands, *Phragmites australis*, Semi-batch, AO7, Artificial aeration, NH_4-N.

1. Introduction

Constructed wetlands for wastewater treatment involve the use of engineered systems that are designed and constructed to utilize natural processes. These systems are designed to mimic natural wetland systems, utilizing wetland plants, soil, and associated microorganisms to remove contaminants from wastewater effluents [1]. Constructed wetlands were mainly used for nutrient and organic matters retention in

Abbreviations

AO7	Acid orange 7
COD	Chemical oxygen demand
DO	Dissolved oxygen
NH_4-N	Ammonium nitrogen
OTR	Oxygen transfer rate
T-N	Total nitrogen
T-P	Total phosphorus
UFCW	Up-flow constructed wetland

domestic and municipal sewage, storm water and agricultural runoff [2-4]. The application of constructed wetlands is gaining in importance as one of the most promising alternatives for the treatment of industrial effluents in developing countries resulted from the transfer of the knowledge, technical collaboration and co-operation by the developed countries [5]. Some researchers have reported the use of constructed wetlands for the treatment of pharmaceuticals and personal care products [6], dyeing wastewater [7, 8], tannery wastewater [9], steel wastewater [10], landfill leachate [11], heavy metals from industrial wastewater [12], seafood wastewater [13] and so on.

Approximately 80 % of the acid and reactive dyes used in textile industrial are azo-compounds, which generally resist aerobic degradation due to the electron-withdrawing nature of the azo linkages. Previous studies have demonstrated that, under anaerobic conditions, azo dyes are easily converted to aromatic amines. However these aromatic amines resist and even inhibit further the anaerobic degradation. Most such end products are further degraded by aerobes [14-16]. Constructed wetlands are simple to use, environmentally friendly, with low construction and operational costs, and efficient enough to treat diverse wastewaters, although the experience in treating textile wastewater is limited [7]. The objective of present study is to evaluate the using of constructed wetland under semi-batch operation for the treatment of azo dye AO7 containing wastewater. A soluble non-reactive acid azo dye, Acid Orange 7, was selected as a model dyeing pollutant in this study because it is widely used in a variety of industries such as textiles, inks, paper, plastics and leather and it is stable to chemical and biological oxidation. The emergent plant selected in our study was *Phragmites australis*. This plant can withstand extreme environmental conditions, including the presence of toxic contaminants such as heavy metals [17, 18].

2. Materials and Methods

Three parallel laboratory-scale constructed wetland reactors (A, B and C) with 30 cm height and 18 cm diameter were developed. Two wetland reactors (aerated (A) and non-aerated (B)) were planted with *Phragmites australis* and the reactor C was remaining unplanted (control). The wetland reactors were operated in semi-batch basis where the synthetic wastewater was fed to the bottom of wetland reactors by peristaltic pump at flowrate 4.46 mL/min for 4 h. Then, the pumping will be terminated and the aeration for the aerated wetland reactor will be started for 19 h. Two porous air spargers were installed at 15 cm below the media surface of aerated wetland reactor to provide artificial aeration for supporting the growth of aerobic microbes. The wetland reactors were in the settle/idle condition for 45 min and then treated wastewater will be discharged in 15 min from the middle of the wetland reactors (S3). The wetland reactors were operated with 2 days of hydraulic retention time. Figure 1 shows the schematic diagram of an wetland reactor.

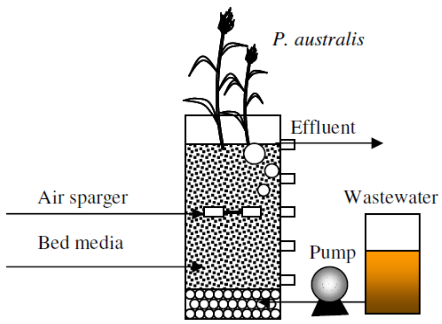

Fig. 1. Schematic Diagram of Constructed Wetland Reactor.

The media used was gravel with average size 6 mm. The media was immerzed in activated sludge which collected from a sewage treatment plant in order for microbes immobilized on the media through attachment. Then, the media was loaded to the reactor and the emergent plant, *Phragmites australis*, was planted about 2-15 cm below the media surface. These wetland reactors were designed with five water sampling points which were at 30 (S1), 23 (S2), 16 (S3), 9 (S4), and 2 cm (S5) from the bottom of the wetland reactor. The wetland reactor and operation characteristics are summarized in Table 1.

Table 1. Wetland Reactor Characteristics.

Characteristic	Value
Total column height	30 cm
Column diameter	18 cm
Height of gravel bed	28 cm
Volume of gravel bed	7.12 L
Average gravel size	6 mm
Average gravel bed porosity	30 %
Average void volume of gravel bed	2.14 L
Hydraulic retention time	2 d

During textile processing, concentrations of AO7 in wastewaters of 10 up to 80 mg/L have been reported [19]. Thus, the synthetic wastewater used containing 50 mg/L of AO7 and composed of a base mixture of carbon sources, nutrients and buffer solution. The composition of wastewater was giving 326 mg/L COD, 62 mg/L T-N and 5 mg/L T-P. The influent and effluent samples were collected daily and were analyzed to examine the treatment performance of the wetland reactors. COD and NH_4-N were determined by using the HACH DR 2800 colorimetric method. The concentration of the AO7 and aromatic amines was determined by the absorbance at wavelength of 480 nm and 245 nm, respectively, using an UV-VIS spectrophotometer (U-2810 spectrophotometer). Dissolved oxygen (DO) was measure using a DO meter (YSI 5000).

3. Results and Discussion

3.1. DO monitoring

The oxygen content in wetland reactor plays an important role in the biodegradation of organic pollutants. Based on the DO monitoring (Fig. 2), it was observed that the DO in effluent from the aerated wetland reactor was in the ranges 2.5-4.5 mg/L whereas in the non-aerated wetland reactor, it ranges 0.5-2 mg/L. The DO in the control wetland reactor was below 0.5 mg/L. This shows that the artificial aeration increased the DO in the wetland reactor and it can support the growth of aerobic microbes for organic matters removal.

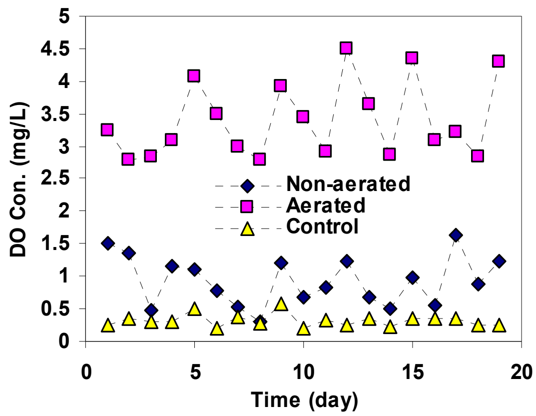

Fig. 2. DO Monitoring for the Effluent from Aerated, Non-aerated and Control Wetland Reactors.

The higher DO in the non-aerated wetland reactor compared to the control wetland reactor shows the important of emergent plant to improve the oxygen level in the wetland reactor. The main role of wetland vegetation is attributed to the modification of soil texture, hydraulic conductivity and soil chemistry by the growth of plant roots and rhizomes. Other relevant features are the release of exudates (vitamins and antibiotics, among others) to the rizosphere and oxygen (for *P. australis*, 0.02–12 g O_2 m^{-2} day^{-1}), which oxygenate the substrate, via direct atmospheric diffusion, thus creating aerobic degradation zones within the bed. Oxygen transfer rates (OTR) have been calculated for plant roots, giving values of 0.02–12 g O_2 m^{-2} day^{-1} for *P. australis* [20, 21]. This suggests the contribution of the emergent plant on oxygen supply through their root systems into the wetland bed in macro-scale. A significant portion of oxygen needed to support aerobic degradation processes in wetlands is obtained through lacuna translocation from the atmosphere to the rhizosphere by rooted aquatic macrophytes [22, 23].

3.2. Emergent plant monitoring

Figure 3 shows the photos of transplanted *P. australis* in the wetland reactors before and after the addition of AO7. Toxic signs like chlorosis symptoms and leaf anatomical changes were observed in the *P. australis* after the addition of

AO7 into the wetland reactors. From the earlier observations, almost all of the leaves in the *P. australis* in the aerated and non-aerated wetland reactors turned to yellowish and no new stem was found after the addition of 50 mg/L AO7. However, after the fourth week, the *P. australis* seems to be able to adapt to the synthetic azo dye containing wastewater and the number of stem was increased as the operation continue. It was found that the emergent plant growth better in the aerated wetland reactor than the non-aerated one. The emergent plant *P. australis* has been shown to survive and reproduce well in the aerated reactor than the non-aerated reactor after the addition of AO7. The toxic signs of plants in the non-aerated up-flow constructed wetland reactors (UFCW) could due to the effects of intermediate products generated through the reduction of AO7 [24].

Fig. 3. *Phragmites Australis* Monitoring in Wetland Reactors.

3.3. Treatment performance in wetland reactors

The influent of the synthetic wastewater contains about 450, 50 and 35 mg/L of COD, AO7 and NH_4-N, respectively. As shown in Table 2, the COD removal efficiency in aerated and non-aerated wetland reactors was 95 and 62 %, respectively. The artificial aeration in the aerated wetland reactor boosted the biodegradation of organic matters. The aerobic condition developed at the aerated wetland reactor facilitated the growth of aerobic microbes and enhanced the biodegradation of organic matters that comprised of substrates and aromatic amines. Besides, the higher COD removal efficiency in the non-aerated wetland reactor than the control wetland reactor shows the presence of *P. australis* increased the COD removal. The *P. australis* transfer the oxygen from the atmosphere to the wetland bed through their roots. This was shown in higher DO level in the non-aerated wetland reactor than the control wetland reactor in Fig. 2. Emergent plants can contribute to wastewater treatment processes in a number of ways, such as settlement of suspended solids, providing surface area for microorganisms and providing oxygen release [21, 25]. The roots provide a huge surface area for attached microbial growth, and plants can also facilitate aerobic degradation by releasing oxygen to the rhizosphere, but oxygen release rates are difficult to quantify and the overall effect on pollutant removal is probably varying [21, 26].

As shown in Table 2, the AO7 removal efficiency for all wetland reactors was higher than 94 % which shows the ability of the semi-batch operated wetland reactors in removing azo dye. It was found that the aerated wetland reactor performed slightly better than the non-aerated wetland reactor. Generally, bacterial azo dye biodegradation proceeds in two stages. The first stage involves reductive cleavage of the dyes' azo linkages, resulting in the formation of - generally colorless but potentially hazardous - aromatic amines. The second stage involves degradation of the aromatic amines. Azo dye reduction usually requires anaerobic conditions, whereas bacterial biodegradation of aromatic amines is an almost exclusively aerobic process [27, 28]. A wastewater treatment process in which anaerobic and aerobic conditions are combined is therefore the most logical concept for removing azo dyes from wastewater [29-31]. As shown in Fig. 4, the level of aromatic amines in the aerated wetland reactor was far lower than the non-aerated wetland reactor. This indicates the aerated wetland reactor not only able to remove color but also the aromatic amines generated from the reduction of azo dye. The aromatic amines may accumulate in the non-aerated and control wetland reactors and contribute to higher COD in effluent.

Table 2. Treatment Performance of Constructed Wetland.

Parameters	Influent (mg/L)	Reactors	Effluent (mg/L)	Removal (%)
COD	457.1 ± 17.6	Control	220.8 ± 11.4	51.7
		Aerated	21.8 ± 15.6	95.2
		Non-aerated	174.5 ± 12.4	61.8
AO7	50.6 ± 3.6	Control	2.9 ± 0.6	94.3
		Aerated	0.2 ± 0.2	99.6
		Non-aerated	2.3 ± 1.1	95.4
NH_4-N	35.3 ± 15	Control	33.9 ± 5.7	3.9
		Aerated	4.9 ± 2.2	86.1
		Non-aerated	30.2 ± 4.6	14.4

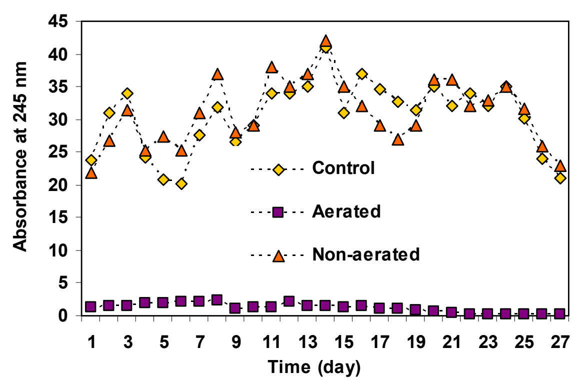

Fig. 4. Aromatic Amines Monitoring in Wetland Reactors for Treating 50 mg/L Acid Orange 7-Containing Wastewater.

Nitrification and denitrification are the main processes for nitrogen removal from wastewater. Denitrification is an anaerobic heterotrophic microbial process often limited by the presence of oxygen (O_2) and the availability of labile carbon substrates. Nitrification is an aerobic chemoautotrophic process. Thus, oxic and anoxic microzones must be coupled in space and labile C abundant for optimal microbial transformation and removal of water soluble N-compounds [32, 33]. As shown in Table 2, the NH_4-N removal efficiency in the aerated wetland reactor

(86 %) was significantly higher than the non-aerated wetland reactor (14 %). In order to enhance N removal efficiency, oxygen must be provided to the nitrifying microbes through oxygenation of the wetland matrix via plant presence, artificial aeration or other means (e.g., diffusion). Artificial aeration requires energy input and additional cost, but in some instances, it may still be profitable. On the other hand, it was observed that the non-aerated wetland reactor performed better in the removal of NH_4-N than the control reactor. This shows the presence of *P. australis* enhanced the removal of NH_4-N. A lower N removal rate in control treatment was not surprising for unplanted-bed wetland treating high N polluted water. Several experimental studies on N removal in treatment wetlands confirmed that unplanted treatment had a lower N removal compared with planted treatment for many cases [34-36]. The NH_4-N can be uptake by the emergent plant and/or degraded by the nitrifying microbe growth near the root of the plant where the oxygen can be obtained to support the nitrification process.

4. Conclusions

Our semi-batch operated wetland reactors planted with *P. australis* show the potential for treating azo dye containing wastewater. Artificial aeration and the presence of *P. australis* had a significant impact on the removal of organic matters, AO7, aromatic amines and NH_4-N. Above 94 % removal of AO7 was observed in all wetland reactors in present study. The aerated wetland reactor with higher oxygen level performed better in the removal of organic matters, aromatic amines and NH_4-N compare to other wetland reactors. In the aerated wetland reactor, the aerobic and anaerobic conditions was developed at the upper bed and lower bed, respectively, due to the artificial aeration by the installment of two porous air spargers at 15 cm below the media surface. The aerated wetland performed better than the non-aerated wetland in terms of the removal of organic matters and NH_4-N and it shows a promising approach to enhance the performance of the wetland reactor.

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