Wastewater Treatment Using Up-flow Constructed Wetland

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1. Introduction

A number of azo dyes are widely used as colorants in industries such as textiles, paper, plastics, cosmetics, and leather. Their highly colored effluents' disposal into the environment can be extremely deleterious. The removal of azo dyes from effluents is important due to their mutagenicity and carcinogenicity together with their intense coloration.

Constructed wetlands are nowadays a well-accepted ecofriendly wastewater treatment technology because of it's utilization of natural processes, simple construction and low maintenance. These systems are based on the functioning of natural ecosystems and the treatment processes involve complex interactions between soil, water, plants and microorganisms which are generally efficient in the removal of organic matter (BOD/COD) and suspended solids (SS), however, the removal of nitrogen and phosphorus is often relatively poor. Treatment of wastewater in constructed wetland systems includes biological and biochemical processes.

In order to utilize constructed wetland systems in treating dyeing wastewater efficiently, the incorporation of anaerobic followed by aerobic conditions is necessary for the wetland reactor. As a result, an up-flow constructed wetland (UFCW) is used here with the bottom reactor in an anaerobic condition for decolorization and denitrification, and the upper layer in aerobic condition for biodegradation of organic pollutants, nitrification and mineralization of aromatic amines. Supplementary aeration is applied to the UFCW reactor to control the ratio of aerobic and anaerobic regions more effectively for the removal of color, organic matters, nutrients and aromatic amines. Azo dye Acid Orange 7 (AO7) is used as a model molecule because it is a non-reactive, acidic azo dye widely deployed in the textile industry and to a lesser extent in paints, inks, plastics and leather. Due to the non-degrading nature of its intermediate products, the COD concentration is persistent in the effluent. Since aerobic biodegradation is capable of mineralizing these intermediate products, it is possible that combined anaerobic–aerobic treatment can efficiently remove color and at the same time completely degrade organic matter. Hence, UFCW reactors with supplementary aeration is proposed in this study for the removal of color, aromatic amines and nutrients. The aim of which is to examine and compare the treatment performance of Acid Orange 7 (AO7) containing wastewater by up-flow constructed wetland (UFCW) at different AO7 concentrations, hydraulic retention times (HRT) and alternative of supplementary aeration.

2. Constructed Wetland Methods

Both aerated and non-aerated, parallel laboratory-scale (UFCW) reactors are constructed to study the treatment of AO7

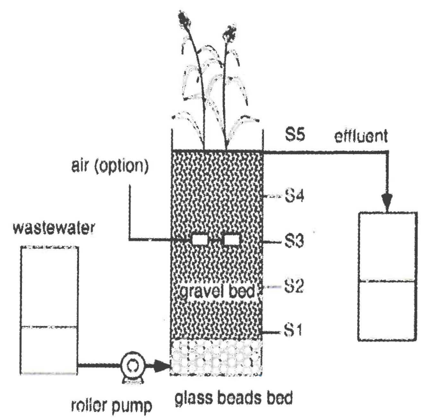

Figure 1. Schematic Diagram of an UFCW

containing wastewater. The reactors, located indoor, are temperature stabilized at 23 ± 3 °C. Fig. 1 shows the schematic diagram of the up-flow constructed wetland reactor, the size of which is 18 cm in diameter and 70 cm in height, and filled with 5 mm glass beads to a depth of about 6 cm at the bottom to ensure uniform distribution of influents. Gravel with an average diameter of 5.7 mm is filled above the glass beads bed and acts as a medium to support the growth of emergent plants and biofilms. The reactors are designed with five water sampling points along the reactors as shown in Fig. 1, where the water level is maintained the same as that of surface gravel bed. In the aerated reactor, supplementary aeration is introduced at 30 cm below the bed surface through two porous air spargers. The macrophyte selected for this study is P. australis with its plant shoots of P. australis planted about 2-15 cm deep into the gravel medium. After transplantation, the wetland reactors are fed with tap water and seeded with a small amount of activated sludge for biofilm to be established. Synthetic wastewater is then designed and used to simulate to the characteristics of azo dye-containing wastewater. The synthetic wastewater consists of organic carbon, nutrients and buffer solution of the following composition (concentration in mg/L): C6H5COONa (107.1), CH3COONa (204.9), NH4NO3 (176.1), NaCl (7.0), MgCl2_6H2O (3.4), CaCl2_2H2O (4.0) and 2HPO4_3H2O (36.7) giving CODCr 326 mg/L, T-N 62 mg/L, and T-P 5.0 mg/L. The concentrations of azo dye AO7 employed in this study are 50 and 100 mg/L. The average inlet wastewater pH is about 7.7 and both influent and effluent water samples of the wetland reactors are taken in 3 periods for a year to gauge their performances. In the first 3 months the reactors are fed with synthetic wastewater and the hydraulic retention time is maintained at 3 d based on average void volume of gravel bed, at an AO7 concentration of 50 mg/L. During the following 6 months, AO7 concentration is increased to 100 mg/L with other factors constant to study the effect of AO7 concentration on the in terms of bio-oxidation of organic matters, decolorization, nitrification and denitrification. In the last 4 months hydraulic retention time is increased to 6 d and AO7concentration is set at 100 mg/L. Influent and effluent water samples are then analyzed to examine the treatment performance of the wetland reactors.

3. Results and discussion

3.1. Oxidation Reduction Potential (ORP) and Dissolved Oxygen (DO) profiles.

The aerobic and anaerobic conditions in a wetland reactor can be distinguished by its ORP and DO profiles. Constructed wetlands for wastewater treatment can be designed to favour a wide range of redox conditions, enhancing a variety of biological processes and removal of multiple pollutants in the same wetland bed. ORP and DO monitoring observation show aerobic and anaerobic conditions developed at the upper and lower beds, respectively, in the aerated wetland reactor. Almost all the non-aerated wetland reactors are found to be in anaerobic condition. Fig. 2 shows that in the wetland reactor with supplementary aeration, the ORP and DO above aeration points are in the range of 6–252 mV and 0.6–7.0 mg/L, respectively. The decrease in DO and ORP from 2.8 mg/L and 235 mV to 0 mg/L and 155 mV, respectively, along the media bed of the non-aerated wetland reactor shows development of anaerobic conditions in the whole media bed except at the top layers. The aerobic and anaerobic regions would influence the activity of microbes in the reduction of azo bond of AO7, degradation of organic substrates and intermediate aromatic amines, nitrification and denitrification.

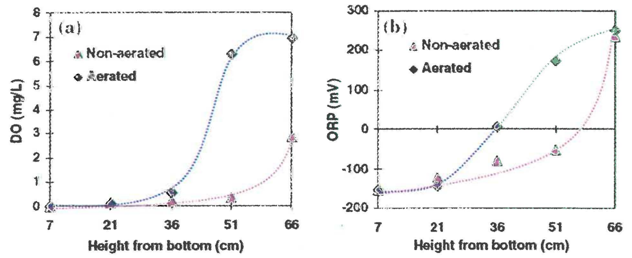

Figure 2. DO and ORP profiles in UFCW reactors

3.2. Treatment performance of UFCW reactors

During 1 year monitoring, the effects of different AO7 concentrations, HRT and alternative supplementary aeration on the performance of UFCWs have been evaluated. Generally, the aerated reactor outperformed the non-aerated reactor for removal of organic matters, NH4-N and aromatic amines, generated from the anaerobic degradation of azo dye AO7. On the other hand, non-aerated wetland reactors performed better in removing color and NO3-N because anaerobic microbials play the main role in the removal of these pollutants. The AO7 removal efficiency in the non-aerated wetland reactor maintained at about 96% whereas in the aerated wetland reactor it dropped drastically as the AO7 concentration increased from 50 to 100 mg/L. This may ascribe to the limitation of the removal capacity of a wetland reactor and/or shock loading which may cause inhibitory effects on the microbial activities. During operation at 6 d of HRT, AO7 removal efficiency in both reactors are not significantly different with 100 mg/L of influent AO7. Influent and effluent concentrations and removal statistics for COD, T-N, T-P, NH4-N, NO3-N and AO7 are summarized in Table 1. The removal of AO7, organic compounds, NH4-N and NO3-N are mainly due to microbes in the reactors because the contribution of adsorption for these pollutants by gravel has been proven negligible. On the other hand, T-P removal is contributed by adsorption, precipitation and uptake by the emergent plant and biomass. T-P removal efficiency dropped gradually due to saturation of adsorption capacity, where its removal efficiency for both reactor types are not much different. Supplementary aeration showed no significant effect on T-P removal. As the influent AO7 concentration increased from 50 to 100 mg/L, COD removal efficiency and nitrification in the non-aerated wetland reactor, and denitrification in the aerated wetland reactor decreased to some extent.

3.3. COD removal in UFCW reactors

Organic compounds degrade both aerobically and anaerobically by heterotrophic microorganisms in the wetland systems depending on the oxygen concentration in the bed, which for aerobic degradation, can be supplied by diffusion, convection and oxygen leakage. 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. COD removal efficiency in the aerated wetland reactor is better than in the non-aerated wetland reactor. Supplementary aeration could thus be used in addition to macrophytes further reduce the organic contaminants in wastewater. Reduction in COD removal efficiency in the non-aerated wetland reactor is mainly attributed to the accumulation of intermediate aromatic amines through partial AO7 degradation, confirmed via absorbance at 248 nm. The generated aromatic amines that were not degraded in the non aerated wetland reactor contributed to higher COD in effluents compared to the aerated wetland reactor, where the aerobic condition at the upper bed of reactor facilitated the degradation of these intermediate products. The aerobic condition at the upper bed of the aerated reactor showed better degradation of organic matters, contributed from sodium benzoate, sodium acetate and aromatic amines, compared to the mostly anaerobic condition in the non-aerated wetland reactor.

HRT is an important operating parameter in various biological systems including constructed wetlands where lower HRT results in higher organic loading rates, causing a reduction of reactor volumes required to achieve a specified removal performance. As expected, COD removal efficiency improved slightly for both reactors as the HRT increased from 3 to 6 d, since higher HRT implies smaller loading rate and thus, a higher removal efficiency. Long contact time between the microbial attached on the media bed and root of emergent plant increases removal efficiency of organic matters. However, a HRT of 3 d is adequate to result in an acceptable removal efficiency of organic matters. Fig. 3 shows the COD profile in the aerated and non-aerated wetland reactors for the treatment of AO7 containing wastewater at different AO7 dosages and HRT. The trend of COD removal is different between the aerated and non-aerated wetland reactors.

COD concentrations from the bottom to the top of non-aerated wetland reactors became higher as the AO7 concentration increased from 50 to 100 mg/L. The higher COD is due mainly from the AO7 and intermediate aromatic amines. In the aerated reactor, the COD concentrations at the lower bed are higher as the AO7 concentration increased, whereas are close to each other at the upper beds. As HRT increased from 3 to 6 d, COD concentrations become lower in the non-aerated reactors. The increase of HRT from 3 to 6 d improved the biodegradation of organic matters along the reactor as the contact time between biofilm and wastewater became longer. In the case of aerated reactors, the COD at the upper bed is not much different. The artificial aeration at the upper bed could maintain the COD removal efficiency despite the increase of HRT and AO7 concentration.

3.4. AO7 removal in UFCW reactors

AO7 removal efficiency in both reactors are almost the same and > 94% for both AO7 concentrations tested and at different HRT. At 50 mg/L of AO7, its removal efficiency is higher than 95% within the same period of operation because the area of anaerobic conditions at the lower bed of both reactors are sufficient for anerobic microbial action. As AO7 concentration increased to 100 mg/L, the non-aerated reactor is able to maintain the same removal efficiency but a drastic drop is observed in the aerated wetland reactor. This situation recovered as the AO7 removal efficiency improved to 95% in a few months, attributed to the different ratio of anaerobic and aerobic regions in both reactors. In the non-aerated wetland reactor, almost the entire bed was in anaerobic condition, except at the top layer of the reactor. The bigger anaerobic region in the non-aerated reactor facilitated the removal of color. The anaerobic lower bed region of the aerated reactor is insufficient for 100mg/L AO7 reduction in the early stage, due to the overload of the biodegradation capacity of the lower bed of aerated reactor. However, with time, the increasing mass of anaerobic microbes could be one of the reasons for achieving 95% of AO7 removal efficiency. As shown in Table 1, AO7 removal efficiency increased slightly in both reactors as HRT increased from 3 to 6 d. The average AO7 removal efficiency for both reactors are 98% which shows the high performance of the UFCW in the treatment of azo dye AO7 containing wastewater.

The molecular structure of azo dye AO7 consists of an azo bond, a benzene ring, and a naphthalene ring, each exhibiting different absorbance peaks, in the visible region (480 nm) for the former, and in the UV region for the latter two. AO7 biodegradation severs the azo double bond, reducing the absorbance at 480 nm and producing aromatic amines such as sulfanilic acid (SA) and 1-amino-2-naphthol (1A2 N). The incorporation of anaerobic and aerobic conditions in a wetland reactor is important for treatment of azo dye-containing wastewater. The decolorization of azo dye occurs when its bond is broken and the aromatic amines generated can be further mineralized. The aromatic amines accumulated along the non-aerated reactor increased as the AO7 concentration increased from 50 to 100 mg/L, which is also observed at the lower bed of aerated reactor but most of the their mineralization occurred at the upper bed in the aerated reactor.

3.5. Nitrogen removal in UFCW reactors

Nitrogen removal from wastewater, nitrification and denitrification, in the aerated and non-aerated reactors are dominant in nitrification and denitrification, respectively, corresponding to aerobic and anaerobic conditions developed in these reactors. The aerated and non-aerated reactors performed better in the removal of NH4-N and NO3-N, respectively. Both processes are affected by the increase of AO7 concentration in the influent. In the aerated reactor, NH4-N removal efficiency differed slightly but NO3-N removal efficiency dropped tremendously with AO7 increase. The non-aerated reactor shows an almost 100% removal efficiency for NO3-N but dropped to 50% for NH4-N as AO7 increased. The azo dye AO7 and aromatic amines generated may cause partial inhibitory effect on the activity of nitrifying and denitrifying microbes.

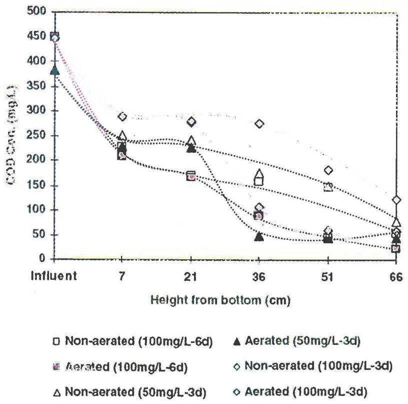

Figure 3. COD profiles in UFCW reactors

The trend of NH4-N concentration along the media bed is different between the aerated and non-aerated reactors, where its tandem drop above the aeration point implicated the merit of supplementary aeration in UFCW reactor. The aerobic condition at the upper bed of the aerated reactor facilitated the nitrification. The slight decrease in NH4-N concentration at the upper bed of the non-aerated reactor could be ascribed to the uptake by P. australis and also the activities of nitrifying bacteria in the rhizosphere. Additionally the increase of AO7 concentration and decrease of HRT caused the NH4-N removal efficiency to drop slightly. In the case of the aerated reactor, NH4-N removal rate is almost similar above the aeration points and achieved > 95% removal efficiency at 51 cm from the bottom, indicating sufficient aeration rate for nitrifying microbes to remove the NH4-N. The anaerobic condition and the presence of carbon sources as electron donors at the bottom of the reactors facilitated denitrification. In the aerated reactor, the higher nitrate concentration above the aeration points corresponds to the decrease of NH4-N concentration through nitrification. Part of the NO3-N generated via nitrification is denitrified in the upper bed of the aerated reactor. The higher NO3-N concentration in the effluent of aerated reactor may be due to the partial inhibition of denitrification and this could be a possible drawback of artificial aeration if excessively high oxygen concentrations are responsible for a combined reduction of denitrification and N removal rates.

Denitrification rates can be enhanced as oxygen content is reduced by reducing the aeration rate. However, this will affect the nitrification rate. In order to remove both NH4-N and NO3-N simultaneously, supplementary aeration must be controlled to enhance the activity of microbes for nitrification and denitrification. Switching the aeration on and off to the upper bed of the reactor will stimulate the nitrification and denitrification processes in this area alternatively. Eventually, both NH4-N and NO3-N can be removed simultaneously in an UFCW reactor.

4. Conclusion

The aerated wetland reactor removed organic matters, NH4-N and aromatic amines more effectively than the non-aerated one. Increasing AO7 concentration deteriorated the biodegradation of organic matters and nitrification in the non-aerated reactor. It also affected the denitrification and decolorization in the aerated reactor. The AO7 and aromatic amines may have inhibitory effect on the activity of nitrifying and denitrifying microbes. As the HRT increased to 6 d, the removal of NH4-N and NO3-N in the aerated reactor is not changed, but NH4-N removal efficiency improved tremendously in the non-aerated reactor.

Table 1. Treatment performance of aerated and non-aerated UFCW reactors

Wastewater	HRT (d)	Parameters	Influent con. (mg/L)	Effluent con. (mg/L)		Removal efficiency (%)	
				Aerated reactor	Non-aerated reactor	Aerated reactor	Non-aerated reactor
50 mg/L AO7	3	COD	383 ± 14	54 ± 16	68 ± 19	86	82
		AO7	51 ± 2	1.1 ± 0.6	2.0 ± 0.6	98	96
		T-N	55 ± 3	18 ± 4	20 ± 3	67	64
		T-P	6.7 ± 0.3	5.0 ± 0.3	4.8 ± 0.5	25	28
		NH_4-N	35 ± 3	1.5 ± 1.1	19.6 ± 4.2	96	44
		NO_3-N	30 ± 2	4 ± 3	0	86	100
100 mg/L AO7	3	COD	450 ± 8	65 ± 18	119 ± 13	86	74
		AO7	103 ± 3	6.2 ± 0.6	3.9 ± 0.6	94	96
		T-N	59 ± 2	27 ± 4	30 ± 2	54	49
		T-P	6.5 ± 0.3	5.5 ± 0.6	5.7 ± 0.5	15	12
		NH_4-N	36 ± 4	0.5 ± 0.4	27.3 ± 5.0	99	24
		NO_3-N	28 ± 2	21.8 ± 5	0.2 ± 0.4	22	99
100 mg/L AO7	6	COD	454 ± 11	44 ± 17	108 ± 13	90	76
		AO7	103 ± 3	2.5 ± 0.3	2.1 ± 0.2	98	98
		T-N	59 ± 2	28 ± 5	33 ± 6	53	44
		T-P	6.5 ± 0.2	5.2 ± 1.1	5.3 ± 0.9	20	18
		NH_4-N	35 ± 3	0.6 ± 0.2	19.6 ± 5.0	98	44
		NO_3-N	28 ± 2	22.1 ± 4.0	1.8 ± 0.6	21	94

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