

Application of A2O moving-bed biofilm reactors for textile dyeing wastewater treatment

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Abstract—A three-stage pilot-scale moving-bed biofilm reactor (MBBRs, anaerobic-anaerobic-aerobic in series) was investigated to treat textile dyeing wastewater. Each reactor was filled with 20% (v/v) of polyurethane-activated carbon (PU-AC) carrier for biological treatment. To determine the optimum operating conditions of MBBRs, the effect of PU-AC carrier, its packing percentage (v/v%) and pH control on COD removal were analyzed by batch experiments. The MBBRs were inoculated with activated sludge obtained from a local dyeing wastewater treatment plant. The MBBR process removed 86% of COD and 50% of color (influent COD=608 mg/L and color=553 PtCo unit) using relatively low MLSS concentration (average 3,000 mg/L in biomass attached to PU-AC carrier) and hydraulic retention time (HRT=44 hr). The MBBR process showed a promising potential for dyeing wastewater treatment.

Key words: Moving-bed Biofilm Reactor (MBBR), Carrier, Decolorization, Dyeing Wastewater, Extracellular Polymeric Substances (EPS)

INTRODUCTION

Textile wastewater discharged from printing and dyeing processes is characterized by a considerable amount of suspended solids and weakly biodegradable substances such as additives, detergents, surfactants and dyes [1]. It exhibits highly fluctuating pH, high temperature and high COD and color concentrations. Stringent government legislation is forcing dyeing industries to treat their wastewater to meet more strict effluent standards. Several methods such as chemical coagulation, electrochemical oxidation, filtration and biological treatment have been developed to treat dyeing wastewater, but most of them could not be used individually in full scale to meet the effluent standards due to variability in dyeing wastewater composition [2,3].

Combining the advantages of suspended growth and biofilm system, the moving-bed biofilm reactor (MBBR) has been developed as one of the most effective processes to treat industrial wastewater because more biomass can be maintained in the reactor by using various types of carriers [4]. Easily biodegradable compounds in the industrial wastewater can be rapidly treated by aerobic treatment, but it is not effective in degrading xenobiotic compounds such as dyes. An anaerobic stage is a very essential phase for biological decolorization [5]. For example, azo dyes can be decolorized by cleavage of the azo bond, with which the color is associated, via anaerobic degradation through the action of non-specific enzymes, and these dyes are reduced as electron acceptors [6]. Degradation possibility of dyes in anaerobic reactor was demonstrated by various researchers [5-7]. However, intermediate products such as aromatic amines can be produced and these are not further degraded in the anaerobic reactor. Seshadri et al. [8] explained that the initial step in the degradation dyes is the cleavage of the azo bond in anaerobic

conditions; however, cleavage is often impossible in aerobic conditions. They observed nearly complete cleavage of the azo dye in anaerobic reactor and the removal of remaining COD to the acceptable limit in sequential aerobic reactor. Thus, anaerobic treatment processes followed by an aerobic treatment would be advantageous in treating the intermediate products such as aromatic amines in dyeing wastewater treatment.

Biofilms are dynamic environments created by extracellular polymeric substances (EPS). The EPS are metabolic products accumulating on the bacterial cell surface that protect bacteria [9]. The EPS matrix sequesters nutrients from the water phase enabling the microorganisms in utilizing optimum available nutrient and carbon. It also supplies photons to organisms located deeper in a microbial mat. In biofilm, EPS provides a framework into which microbial cells are attached for the matrix development. They are also responsible for keeping cells together in the form of biofilms, flocs and sludges [10]. EPS plays a crucial role in creating proper initial conditions for microorganisms to adhere to the polyurethane (PU) surfaces and in speeding up the decolonization process [11]. Therefore, EPS is highly important in biological COD and color removal from dyeing wastewater, especially when the polymer carrier is used [7].

Growth of microbial population is governed by supporting media in the reactor. Selection of supporting media is a determining factor for decolorization of dyeing wastewater. Various researchers demonstrated that PU foam is an excellent supporting medium for biomass. Such outstanding characteristics were observed in a short period of operation [12,13]. PU foam was used as a support matrix to immobilize anaerobic biomass because the PU matrices provide excellent conditions for anaerobic growth and retention of microorganisms [12,14]. SEM analysis of PU media revealed the presence of mainly dense short rods, filamentous on the outer and inside the cavities of foam [11]. Therefore, the use of PU media can provide high degradation efficiency by the formation of high biomass on

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them [15].

The main objective of this study is to find the effectiveness of anaerobic-anaerobic-aerobic (A2O) moving-bed biofilm reactors (MBBRs) for the treatment of dyeing wastewater. The COD and color removals were monitored over 40 days of operation. The results of this study may offer a promising potential of MBBR process for dyeing wastewater treatment without use of any further chemical treatment (e.g., coagulation and flocculation).

MATERIAL AND METHODS

1. Dyeing Wastewater Characteristics

Dyeing wastewater used in this study was obtained from a dyeing wastewater treatment plant located in Daegu dyeing complex, Daegu, Korea. Its average COD, color, T-N, T-P, pH and temperature were 608 mg/L, 553 PtCo unit, 33 mg/L, 3.5 mg/L, 12.5 and 40 °C, respectively.

2. Preparation of PU-AC Foam

Polyurethane-activated carbon (PU-AC) foam carrier was synthesized by slight modification of the procedure previously reported

by Shin et al. [14]. The PU-AC foam carrier was made from Tween 85 surfactant (Aldrich Chemical Co., USA), Hypol 3000 prepolymer (Dow Chemical Co., USA) and activated carbon (AC). Photographs of PU-AC foam carrier are shown in Fig. 1.

Surfactant solution was prepared by dissolving 30 g of Tween

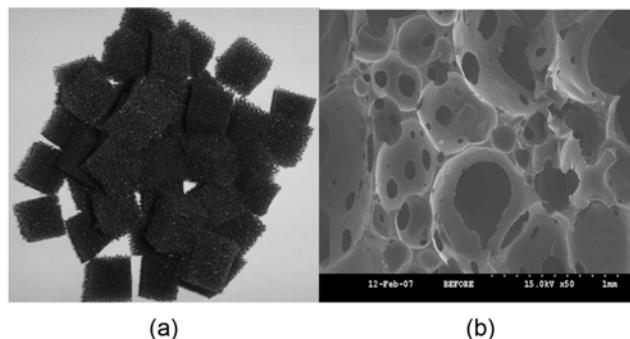


Fig. 1. (a) Photograph and (b) SEM photograph of PU-AC foam (1 cm×1 cm×1 cm, ×50 k magnification, 1 μm).

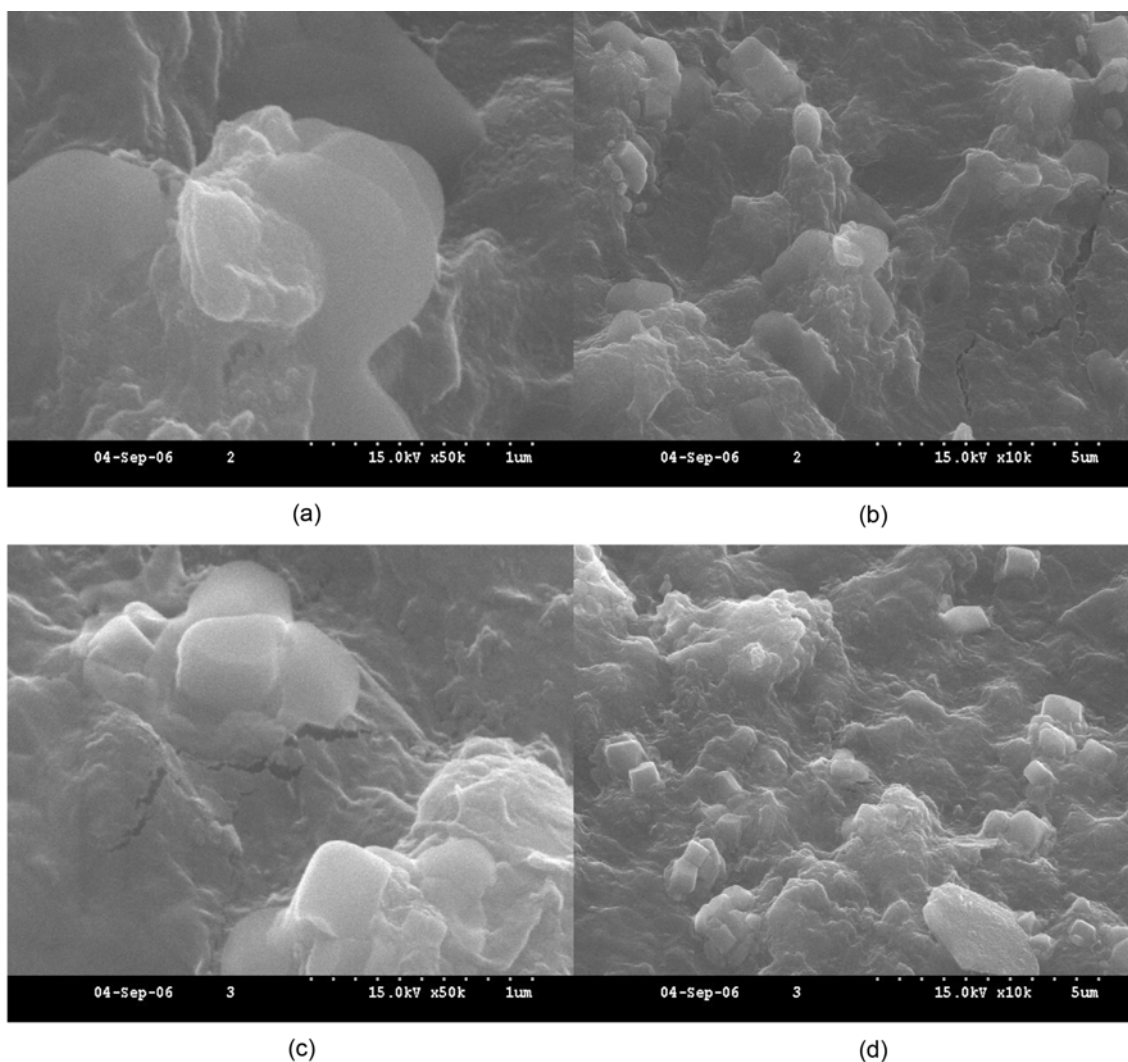


Fig. 2. SEM photographs of microorganisms attached to the polyurethane-AC media surface. (a) Anaerobic (×50 k, 1 μm), (b) Anaerobic (×10 k, 5 μm), (c) Aerobic (×50k, 1 μm) and (d) Aerobic (×10 k, 5 μm).

85 surfactant in 1 L of deionized water. In the surfactant solution, 10 g of activated carbon (AC) was added and cooled to 4 °C in a refrigerator. Hypol 3000 was heated to 55 °C in a water bath and maintained at that temperature for at least 2 hours before being combined with surfactant solution. Approximately 110 g of Hypol 3000 and 110 g of surfactant solution were added into a Teflon beaker (Nalgene, USA), vigorously mixed for 20 seconds using a plastic spoon, and then poured into an 8.5 cm-internal diameter, 12-cm long cylindrical cardboard mold [14]. As the foaming reaction progressed, polyurethane foam expanded to fill the mold. Then, the foam was air dried and the mold was removed. Before further testing, the impermeable "skin" that formed on the outermost layer of the foam (and adhered to the cardboard mold) was removed as were the top and bottom 1 cm of each cylinder. Free surfactant was rinsed from the foam by repeatedly washing with DI water. The PU-AC carrier (1 cm×1 cm×1 cm) had a pore size of 13.52 Å, a surface area of 59.7 m²/g and a density of 1.064 g/m³. The surface of PU-AC media was examined using SEM (scanning electron microscopy, Hitachi S-4300). Photographs of microorganisms attached on the PU-AC carrier are shown in Fig. 2.

3. Pilot-scale MBBR Process

The continuous flow MBBR process (Fig. 3) for dyeing wastewater treatment consists of anaerobic-A, anaerobic-B and aerobic MBBR. The pilot-scale MBBR process consists of three 15 L reactors (work-

ing volume) equipped with a diffused aeration and mechanical stirrers for mixing of wastewater. Each MBBR was filled with PU-AC foam carrier by 20% (v/v). Anaerobic conditions in the first two MBBRs were kept in a dispersed state by a mechanical stirrer and aerobic condition in the last MBBR was maintained by continuous aeration and mechanical stirrer. The pH of the wastewater in storage tank was adjusted to 7 with 1 N of H₂SO₄. Wastewater was fed and discharged by peristaltic pumps (Cole-Parmer, USA) equipped with Easy-load II[®] pump head and MasterFlex[®] Viton tubing. The total hydraulic retention time (HRT) in three MBBRs in series was maintained at 44 h. The pilot-scale MBBR system was operated to treat dyeing wastewater. Before receiving dyeing wastewater, microorganisms were attached to PU-AC foam carrier using activated sludge from a local dyeing wastewater treatment plant located in Daegu, Korea. COD and color removals from dyeing wastewater were tested using batch reactor and pilot-scale MBBRs to investigate the effect of PU-AC carrier addition. The suspension was incubated for approximately seven days to encourage cell growth and the adhesion of freely suspended biomass onto the PU-AC foam carrier.

4. Sample Analysis

Daily wastewater samples were subjected to vacuum filtration by glass microfiber filter (Whatman, GF/C filter, 0.45 µm, 47 mm in diameter). As shown in Table 1, the COD, T-N, T-P and MLSS

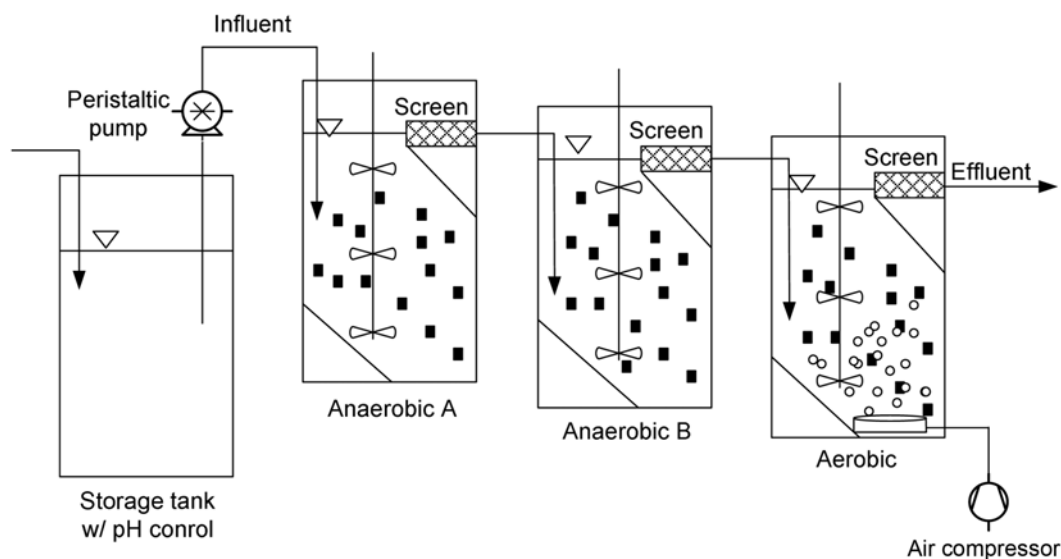


Fig. 3. Schematic of pilot-scale MBBR process for dyeing wastewater treatment.

Table 1. Summary of analytical methods

Parameter	Sources	Apparatus or method
COD	Standard Methods (APHA, 2005)	
Color	-	HP 8452A UV-spectrophotometer
pH	-	Orion 290A pH meter
DO	-	Orion 810 DO meter
T-N	Standard Methods (APHA, 2005)	HP 8452A UV-spectrophotometer
T-P	Standard Methods (APHA, 2005)	Ascorbic acid method
ORP		Orion 290A ORP meter
Biomass	Im et al. [17]	-

concentrations were measured using standard methods [16], and the amount of biomass in PU-AC was measured using the same protocol as Im et al. [17]. Extracellular polymeric substances (EPS) in PU-AC carrier were extracted using EDTA method [18]. The total quantity of extracted EPS was measured by the weight of solids after lyophilization. The protein content in EPS was measured by the modified Lowry method [19].

RESULTS AND DISCUSSION

1. Effect of PU-AC Carrier on COD Removal

Two batch reactors with and without PU-AC media were operated to estimate the effect of PU-AC media addition on COD removal efficiency (Fig. 4). In both anaerobic and aerobic reactors, the COD removal rate was slightly higher in the presence of PU-AC media. Within the first day, 48% and 33% of COD was removed in anaerobic reactors with and without the PU-AC media, respectively (Fig. 4(a)). Approximately 73% and 63% of the total COD was removed during eight-day operation. The amount of COD removed within the first day was 65% and 52% of the total amount of COD removed over eight days. This indicates that a large amount

of easily biodegradable COD fraction was removed within the first one day. The COD removal efficiency with the PU-AC media was higher than that without the PU-AC media because higher amount of biomass was contained in the PU-AC media and the fluidization of the moving media causes high opportunity of contact with the wastewater.

Fig. 4(b) shows that 67% and 57% of COD was removed within the first one day in aerobic reactors with and without the PU-AC media, respectively. Approximately 86% and 75% of the total COD was removed during eight-day operation. This indicates that most of biodegradable COD was removed within the first one day under aerobic conditions and the COD degradation rate under aerobic conditions was faster than that under anaerobic conditions.

2. Effect of PU-AC Media Packing Percentage (v/v%) on COD Removal

The effect of PU-AC carrier packing percentage (10, 20 and 30%, v/v) on the COD removal was also investigated in batch MBBRs. The ratio of COD concentration at time t versus initial COD concentration, $C(t)/C_0$ ratio, is presented in Fig. 5.

Within one day, $C(t)/C_0$ rapidly decreased from 1.0 to 0.76, 0.64 and 0.46 at 10, 20 and 30% of PU-AC carrier packing percentage

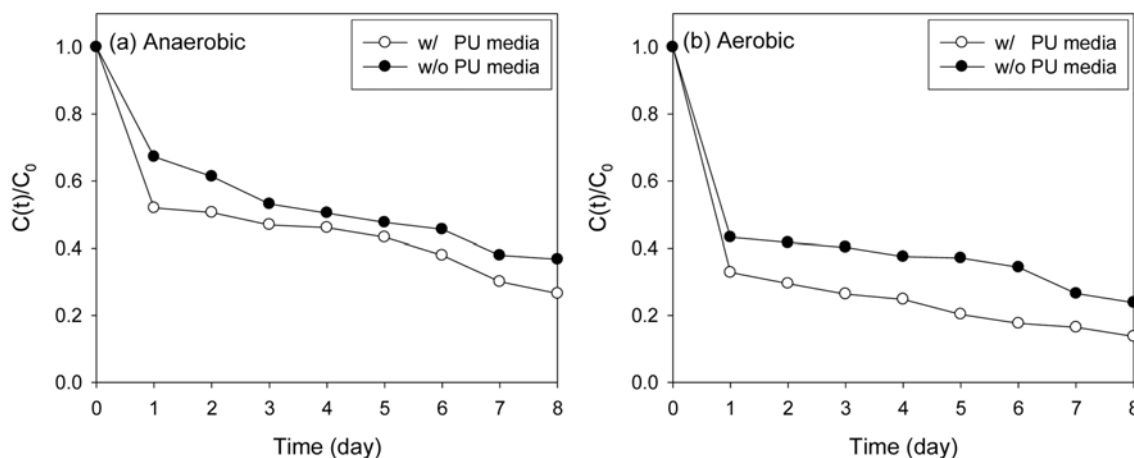


Fig. 4. Effect of PU-AC foam carrier addition on COD removal in batch reactor. (a) anaerobic and (b) aerobic (MLSS=90 mg/L and PU-AC carrier content=20%, v/v).

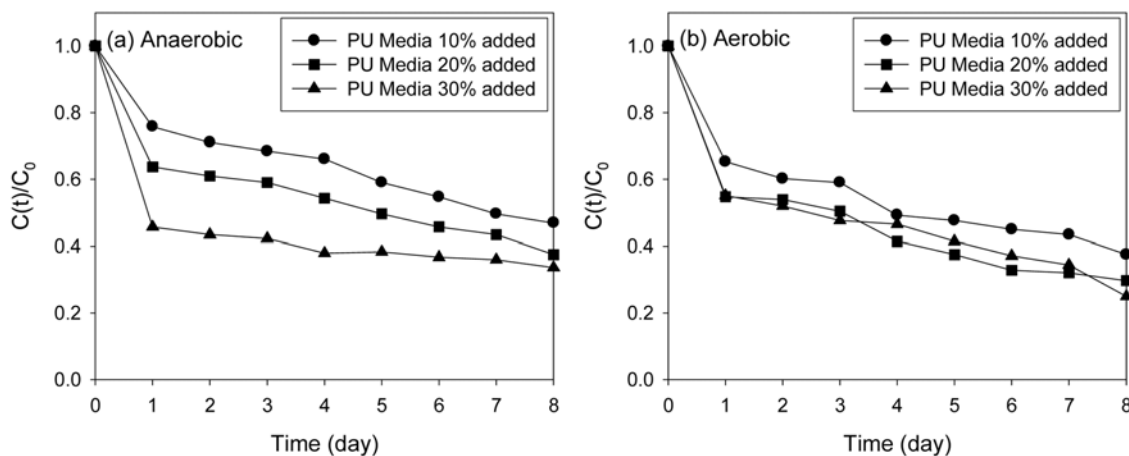


Fig. 5. Effect of volumetric packing percentage of PU-AC carrier on COD removal.

in the anaerobic reactor and to 0.65, 0.55 and 0.55 in the aerobic reactor, respectively. The COD removal within the first one day was 46, 58 and 82% of total COD removal over eight days in the anaerobic reactor at 10, 20 and 30% of packing percentage, respectively; however that in the aerobic reactor was 55, 64 and 60%, respectively. These results indicate that COD removal was enhanced with increasing the packing percentage of PU-AC media from 10% to 30% in the anaerobic reactor and from 10% to 20% in the aerobic reactor, respectively. However, COD removal in the aerobic reactor was not further enhanced as the packing percentage of the media increased from 20% to 30%. Therefore, the optimum packing percent of PU-AC in the anaerobic and aerobic reactors was 30 and 20%, respectively. In addition, the COD removal under anaerobic condition was slower than in aerobic conditions at the same packing percentage except for 30% because a large amount of dyestuff was converted to biodegradable aromatic amines by the cleavage of the dye bond.

After one day, all $C(t)/C_0$ ratios were gradually declining in both the anaerobic and aerobic reactors as shown in Fig. 5. In anaerobic reactors packed with 10, 20 and 30% of PU-AC, 29, 26 and 12% of COD was removed, respectively, compared to that within the first one day. However, any relationship between packing percentage of the media and slow COD removal rate after one day was not found.

3. Effect of pH Control on COD Removal in MBBR

The pH has significant effect on the COD reduction in dyeing wastewater. Most bacteria can grow at around neutral condition (pH 6-8); however, very few species of bacteria can grow under high pH such as raw dyeing wastewater. Therefore, it is essential that the pH should be adjusted to neutrality. Fig. 6 depicts the comparison in the $C(t)/C_0$ profiles between pH 7 (with pH adjustment) and pH 12 (without pH adjustment). pH 7 was better than pH 12 in COD removal from dyeing wastewater. COD was reduced sharply in the first two days at pH 7, while at pH 12 the reduction was gradual.

4. Pilot-scale MBBR Process for Dyeing Wastewater Treatment

The pilot-scale MBBRs in series (anaerobic-anaerobic-aerobic, A2O) were operated to treat real dyeing wastewater (Fig. 7). The removal efficiency of the pilot scale MBBR process is summarized in Table 2.

The first two anaerobic MBBRs removed 78% of COD and 50%

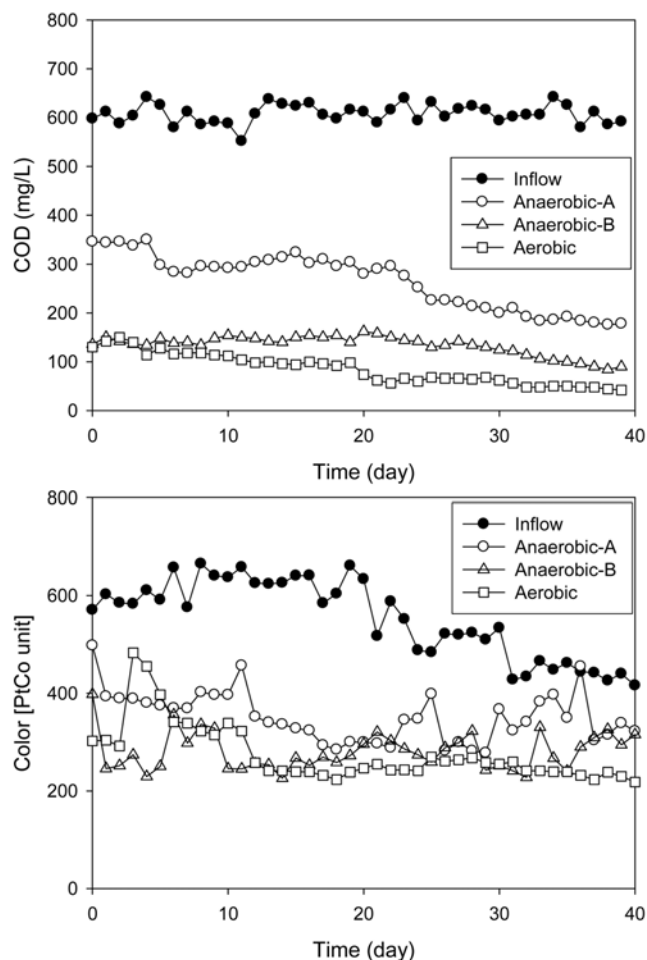


Fig. 7. COD and color removal from dyeing wastewater in pilot-scale MBBR process.

of the color. Most of COD was removed in the anaerobic MBBRs and the remaining COD was further removed by the subsequent aerobic MBBR. However, color removal efficiencies at anaerobic-A, anaerobic-B, and aerobic MBBRs were 37, 13 and 1%, respectively, indicating color removal occurred in only anaerobic MBBRs, not in aerobic MBBR, because dyestuff transformation to aromatic

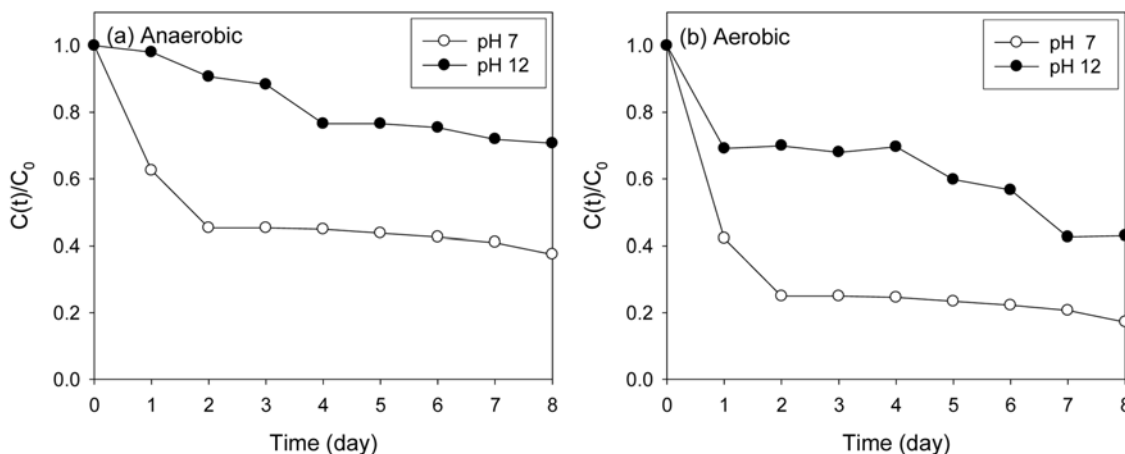


Fig. 6. Effect of pH on COD removal (PU-AC carrier content=20%, v/v).

Table 2. COD and color removal in the Pilot-scale MBBR process

	Influent	Anaerobic A	Anaerobic B	Aerobic	Overall
COD (mg/L)	522-642 (608)*	176-350 (265)	84-162 (134)	42-150 (85)	
Removal rate (%)	-	41-71 (56)	13-35 (22)	-1-16 (8)	74-93 (86)
Color (PtCo unit)	416-666 (553)	278-498 (351)	226-398 (280)	218-483 (275)	
Removal rate (%)	-	-3-55 (37)	-8-37 (13)	-37-23 (1)	17-64 (50)

*Number in the parenthesis indicates the average value

amines occurred under only anaerobic conditions. After the effluent was passed through the last aerobic MBBR, the COD removal efficiency increased to 86% (COD in effluent=42-150 mg/L). In our previous study, 29% of COD and 55% of color were removed in an anaerobic MBBR [14] at which the influent COD and color concentrations were higher (influent COD=807.5 mg/L and Color=3,400 PtCo unit) than those in this study. The discrepancy in removal efficiency is attributed to the difference in the characteristics of applied dyeing wastewaters. Sequential treatment of the anaerobic effluent in the aerobic reactor removed the additional COD, mainly due to the removal of aromatic amines. As summarized in Table 2, COD and color concentrations in the effluent from aerobic MBBR were 85 mg/L and 275 PtCo units, respectively.

Color removal efficiency of the first two anaerobic MBBRs in series was 50%, while that of the aerobic MBBR was 1% only. This means that the most of the color was efficiently removed within the two anaerobic MBBRs. In terms of color removal, after the aerobic MBBR the color removal efficiency was not changed although a small amount of COD was removed as shown in Fig. 7; color removal was mainly achieved at the initial stage of anaerobic treatment. This result was in agreement with the previous report by Panswad et al. [5], who reported that the color removal in anaerobic reactor was in the range of 68 to 72%.

A conventional activated sludge process generally requires 3,000-4,000 mg/L of MLSS concentration in wastewater treatment. In the MBBR process, however, higher removal efficiency was obtained even at a relatively low MLSS concentration (MLSS in the water phase=90 mg/L). This is because a high concentration of biomass is attached to PU-AC carrier (average MLSS=3,000 mg/L), and

Table 3. Biomass concentration attached to the PU-AC carrier

Parameters	Average
Biomass wt (g)	0.0015
PU foam wt (g)	0.105
Biomass wt/PU foam wt	0.015
MLSS (mg/L)	3000

Table 4. EPS and protein in the biomass attached to the PU-AC carrier

	pH	ORP (mV)	EPS (mg/g-VSS)	Protein (mg/g-VSS)
Anaerobic-A	7.6-8.0	-250-300	318.9	141.5
Anaerobic-B	8.5-8.7	-198-230	318.5	169.8
Aerobic	8.8-9.2	20-40	343.6	150.3

also the contact opportunity between the moving carrier and the pollutants in the wastewaters is increased by fluidization, thereby treating the color and COD more efficiently.

Tables 3 and 4 demonstrate the biomass concentration attached to PU-AC carrier and EPS contained in the carrier, respectively. Biomass attached to the carrier surface was about 1.5% (w/w) of PU media; therefore, the MLSS concentration in the MBBR was 3,000 mg/L (Table 3). These conditions enhance COD and color removal from the wastewater. In our previous study [14], the EPS concentration in the aerobic MBBR (640 mg/g-VSS) was much higher than that in anaerobic MBBR (182 mg/g-VSS). In this study,

Table 5. Comparison of different combined processes for COD removal from textile wastewater

Influent COD	Stage-1	Stage-2	Stage-3	Reference
900 mg/L	Chemical coagulation 600 mg/L (33%)*	Activated sludge treatment 120 mg/L (87%)	Tertiary treatment (filtration) 80 mg/L (91%)	Nicolaou and Hadjivassilis [20]
694 mg/L	Chemical coagulation 391 mg/L (44%)	Electro-coagulation 236 mg/L (66%)	Activated sludge treatment 101 mg/L (85%)	Lin and Peng [21]
824 mg/L	Fluidized biofilm process 206 mg/L (75%)	Coagulation 62 mg/L (82%)	-	Park and Lee [22]
872 mg/L	Fluidized biofilm process 272 mg/L (69%)	Chemical coagulation 100 mg/L (89%)	Electrochemical oxidation 40 mg/L (95%)	Kim et al. [23]
807 mg/L	Anaerobic MBBR 572 mg/L (29%)	Two-phase aerobic MBBRs 115 mg/L (85%)	Chemical coagulation 41 mg/L (95%)	Shin et al. [14]
608 mg/L	Two-phase Anaerobic MBBRs 134 mg/L (78%)	Aerobic MBBR 85 mg/L (86%)	-	This study

*Number in the parenthesis indicates COD removal efficiency (%)

however, the EPS concentration in the aerobic MBBR (343 mg/g-VSS) was almost the same as that in the anaerobic MBBR (319 mg/g-VSS). The COD removal was highly enhanced with the increased EPS concentration in the biofilm in both MBBRs. SEM photographs confirmed that abundant amount of bacterial cell was present in the PU-AC carrier (Fig. 2).

The results of COD removal from dyeing wastewater by several combined processes reported in the literature are compared in Table 5. Nicolaou and Hadjivassilis [20] reported that a chemical coagulation+activated sludge process can remove 87% of COD. Lin and Peng [21] also used a combined system composed of chemical coagulation, electro-coagulation and activated sludge. The COD removal efficiency was 85%. Park and Lee [22] and Kim et al. [23] also reported that COD removal efficiencies were 82 and 89% when combining fluidized biofilm process with chemical coagulation for dyeing wastewater treatment. From these comparisons, the A2O MBBR system is more effective in dyeing wastewater treatment than the other systems because the A2O MBBR system has similar efficiency of COD removal without coagulation compared to the other systems. In our previous study [14], the COD removal efficiency of anaerobic-aerobic-aerobic MBBRs without chemical coagulation was 85%, which is the almost the same COD efficiency as A2O MBBRs in this study and increased to 95% after chemical coagulation with FeCl_2 (see Table 5).

In conclusion, our A2O MBBR system showed better performance in dyeing wastewater treatment than other conventional systems such as chemical coagulation and activated sludge system or fluidized biofilm process and chemical coagulation system, and, if the coagulation process is added to the MBBR system, it will be possible for this system to be more effective dyeing wastewater treatment as shown in the previous study [14].

CONCLUSIONS

The effectiveness of pilot-scale moving-bed biofilm reactors (MBBRs) in dyeing wastewater treatment was investigated. In overall, COD and color removals in the pilot-scale MBBR process were 86% and 50%, respectively. The COD and color reductions mainly occurred in anaerobic MBBRs, while a small amount of COD was removed and color was hardly removed in aerobic MBBR. The A2O (anaerobic-anaerobic-aerobic) MBBR system was highly efficient in textile dyeing wastewater treatment and can satisfy the national guideline of effluent qualities in Korea. Thus, biological treatment composed of anaerobic-anaerobic-aerobic MBBR using PU-AC foam as carrier is a viable technique for dyeing wastewater treatment without using further chemical treatment (e.g., coagulation and flocculation).

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