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Suitability of Membrane Bioreactor for treatment of recalcitrant textile dye wastewater utilising white-rot fungi

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Abstract

The performance of a laboratory scale membrane bioreactor (MBR) — utilizing a mixed microbial community dominated by fungi— for treatment of textile dye wastewater was investigated. A synthetic wastewater containing dye, starch (main contributor to total organic carbon, TOC) and other nutrients was used. Preliminary batch tests confirmed the superior decoloration capacity of pure fungus culture (Coriolus versicolor, NBRC 9791) as compared to that of conventional activated sludge. Simultaneous biosorption and biodegradation was evident in case of the fungus, while mainly biosorption was responsible for decoloration by activated sludge. On the other hand, activated sludge demonstrated comparatively faster TOC removal. Interestingly, stable removal of both color (over 99%) and TOC (over 98%) was achieved in the MBR under a hydraulic retention time (HRT) of 1 day. The difference of reactor-supernatant and membrane-permeate quality substantiated the significant contribution of the membrane to the overall dye removal (biosorption, cake layer filtration, biodegradation).

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Suitability of membrane bioreactor for treatment of recalcitrant textile dye wastewater utilizing white-rot fungi

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Abstract: The performance of a laboratory scale membrane bioreactor (MBR) — utilizing a mixed microbial community dominated by fungi— for treatment of textile dye wastewater was investigated. A synthetic wastewater containing dye, starch (main contributor to total organic carbon, TOC) and other nutrients was used. Preliminary batch tests confirmed the superior decoloration capacity of pure fungus culture (*Coriolus versicolor*, NBRC 9791) as compared to that of conventional activated sludge. Simultaneous biosorption and biodegradation was evident in case of the fungus, while mainly biosorption was responsible for decoloration by activated sludge. On the other hand, activated sludge demonstrated comparatively faster TOC removal. Interestingly, stable removal of both color (over 99%) and TOC (over 98%) was achieved in the MBR under a hydraulic retention time (HRT) of 1 day. The difference of reactor-supernatant and membrane-permeate quality substantiated the significant contribution of the membrane to the overall dye removal (biosorption, cake layer filtration, biodegradation).

Keywords: Dye, Membrane Bioreactor, Textile wastewater, White rot fungi

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Dr. Kazuo Yamamoto is a professor at the Environmental Science Center, University of Tokyo, Japan. He is credited with the introduction of submerged membrane bioreactor process for treatment and reuse of wastewater. Apart from his pioneering contribution in the field of membrane bioreactor, his group works on a broad range of topics including bioremediation of contaminated site, hazardous waste management, waste-to-energy processes, ground water management and urban air quality monitoring.

1. Introduction

Textile wastewater is a complex and highly variable mixture of many polluting substances including dye (Robinson et al., 2001). Due to their synthetic origin and complex structure deriving from the use of different chromophoric groups, dyes are extremely recalcitrant. The release of dye effluent in the ecosystem is a remarkable source of esthetic pollution, eutrophication, and perturbations in aquatic life. The presence of dyes or their degradation products in water can also cause human health disorders such as nausea, hemorrhage, and ulceration of skin and mucous membranes, and can cause severe damage to the kidney, reproductive system, liver, brain, and central nervous system (Kadirvelu et al., 2003).

Several physicochemical decolorization techniques have been reported (e.g. adsorption, membrane separation, advanced oxidation process), none, however, has appeared as a panacea due to high cost, low efficiency and inapplicability to a wide variety of dyes. Biodegradation is an environmentally friendly and cost competitive alternative; but the conventional aerobic treatments have been proved rather ineffective while highly toxic aromatic amines can be formed by reductive fission under anaerobic conditions (Hao et al., 2000; Hai et al., 2007).

Unlike bacteria in conventional activated sludge, aerobic white-rot fungi can degrade wide varieties of recalcitrant compounds including textile dyes by non-specific extracellular enzymes (Wesenberg et al., 2003). However, to date, the large-scale application of fungal decoloration has been impeded owing to the lack of a reactor system capable of coping with the difficulties including slow fungal degradation requiring long hydraulic retention time (Moreira et al., 1998), excessive growth of fungi causing reactor-clogging (Zhang et al., 1999), bacterial contamination destabilizing fungal decoloration (Libra et al., 2003), and loss of the extracellular enzymes and mediators with discharged water (Zhang and Yu, 2000).

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It is interesting to note in this context that in contrast to the conventional activated sludge process, membrane bioreactor (MBR) technology offers several advantages including excellent effluent quality, low sludge production, decoupling of hydraulic retention time (HRT) and sludge retention time (SRT), small foot print and flexibility in future expansion (Visvanathan et al., 2000; Yang et al., 2006). An MBR may overcome the aforementioned limitations of conventional fungal reactors.

This study intends to demonstrate the suitability of MBR for practical application of fungal dye degradation. Preliminary batch tests confirmed the superior recalcitrant dye degradation capacity of white rot fungi in comparison to that of conventional activated sludge, while subsequent long-term operation of a membrane-coupled fungi reactor evidenced the potential of MBR for application of fungal remediation of recalcitrant dye wastewater.

2. Materials and methods

2.1 Microorganism

The white-rot fungus *C. versicolor*, NBRC 9791 obtained from the NITE Biological Resource Center (NBRC), Japan was used for this study. Activated sludge sample collected from Shibaura wastewater treatment plant, Tokyo was utilized for comparison of dye degradation capacity of pure fungi culture and bacteria-dominated conventional activated sludge.

2.2 Properties of the selected dyes

Three water soluble dyes purchased from Sigma Aldrich Co., USA were selected for this study. Figure 1 and table 1 show the structures and properties, respectively, of the selected dyes. Dyes can be classified either according to the chromophoric group (e.g., azo, phthalocyanine etc.) or by their application method (e.g., reactive, direct etc.). The azo dyes are by far the most important class, accounting for over 50% of all commercial dyes (Hunger, 2003). Accordingly, two azo dyes (polymeric and reactive) were selected for this study. The third dye was a metal containing direct dye having phthalocyanine as chromophore. It is worth-noting here that both the direct and reactive dyes are utilized for cellulosic fiber. However, reactive dye molecules contain specific functional groups that can undergo addition/substation reactions with the OH, SH and NH_2 groups present in textile fibers, while direct dyes show high affinity for fibers in the presence of electrolytes.

Insert Fig 1

Insert Table 1

2.3 Synthetic wastewater

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For reactor operation, a nutrient-sufficient synthetic wastewater was prepared by adding dye (100 mg/l) and starch (2 g/l)—two common components in real textile wastewater—along with urea (0.1 g/l) and other nutrients (Hai et al., 2008) into tap water. A significant portion of the poorly soluble starch in the wastewater remained in suspended form. The media for batch tests possessed the same composition; however, starch was in soluble form as the final solution was autoclaved in this case. Also high purity Milli-Q water (Millipore, Japan) was used instead of tap water during batch tests. Batch tests were conducted separately for all the three dyes. Since textile effluent may contain a range of dyes, efficient decoloration of a single dye does not necessarily substantiate the suitability of a decoloration process. However, in order to facilitate monitoring of dye structure-specific decoloration, the MBR performance was assessed by sequentially adding individual dyes in the wastewater. This paper will discuss the performance with Poly S119 dye, which was added first since polymeric dyes are believed to represent the majority of the synthetic dyes (Zheng et al., 1999). The total organic carbon (TOC) concentration of the media with Poly S119 dye was around 945 mg/l, of which only around 5% (as estimated from the difference of TOC of media with and without dye) was contributed by the dye.

2.4 Batch test description

100 ml beakers, containing 50 ml of autoclaved culture media (section 2.3) including 100 mg/l of certain dye, were aseptically inoculated with 0.05 gm (dry wt.) active fungi or activated sludge. The fungus was previously grown within colorless media. In order to ascertain the contribution of biosorption to total decoloration, beakers containing biomass (fungi or activated sludge) inactivated by autoclaving (121°C, 0.2 MPa, 15 min.), were also incubated. With the composition mentioned in section 2.3, the pH of the media stood at 4.5, which is favorable to fungi. In view of the fact that the optimum pH for bacteria and fungi are different, the media pH of the beakers containing bacteria was adjusted to 7 using NaOH solution prior to autoclaving. However, abiotic samples under both the pH were incubated to detect any probable chemical effect of pH on media decoloration. The beakers were loosely covered with aluminium foil and then placed under 28 °C on a shaker (BR-300LF, Taitec reciprocal bio-shaker, Japan) at 80 rpm. The batch tests were carried out in triplicates where each run lasted for 7 days. Samples (0.5 ml) were collected daily, diluted four times with Milli-Q water and then color and TOC removals were measured.

2.5 Membrane module and the bioreactor

A spacer-filled bundle of micro-porous, hydrophilicaly treated polyethylene hollow-fibers obtained from Mitsubishi Rayon, Japan was utilized in this study. The specifications of the module have been outlined in table 2. Pulsed backwash with permeate (flowrate=1.67 ml/s,

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duration=3 s per 10 min) and periodic backwash with NaOCl solution containing 500 mg Cl_2 /l (100 ml/ m^2 membrane surface; twice/ week) were applied. In addition, surface aeration was provided with a specially designed diffuser (intensity= 2.5L air/ min, duration=1min per 30 min). Details about the design principle of the module and the corresponding moderate consumption of chemical and air for cleaning (despite their rather frequent application) have been documented elsewhere (Hai et al., 2008).

Insert Table 2

A cylindrical, PVC bioreactor (working volume = 11.8 L) was used in this study (Figure 2). A diffuser supplied continuous air from the bottom of the reactor with an intensity of 5 l/ min for complete mixing and supply of dissolved oxygen to the microbes. The temperature of the reactor was controlled at 29±1°C. It was initially inoculated with pure fungi culture but was operated under an HRT of 1 day without any specific control of bacterial contamination. The MBR was operated continuously for 3 months. Color and TOC in membrane-permeate were measured on every third day while reactor-supernatant samples were analyzed at an interval of 10 days.

Insert Figure 2

2.6 Analytical methods

Color measurements were carried out using a spectrophotometer (U-2010, Hitachi, Japan). The absorbance at the peak wavelength of the dye in consideration was measured to ascertain the extent of media decoloration. Percentage dye removal was calculated using an absorbance vs. concentration calibration curve. TOC was measured with a Total Organic Carbon analyzer (TOC-V, Shimadzu, Japan). The mixed liquor suspended solids (MLSS) concentration was measured according to the standard methods (APHA/AWWA/WEF, 1998).The relative abundance of fungi/bacteria in MLSS was estimated following the method of Jasti et al.(2006) using 20 times diluted samples. Transmembrane pressure (TMP), as an indicator of membrane fouling, was continuously monitored using a vacuum pressure gauge (GC 61, Nagano keiki Co. Ltd., Japan).

3. Results and discussion

3.1 Performance comparison of conventional activated sludge and pure fungi

Preliminary batch tests were carried out to confirm the superior dye degradation capacity of the utilized fungus strain. The fungal dye degradation capacity was compared with that of bacteria-dominated conventional activated sludge. It is worth-reiterating here that

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decoloration was monitored in flasks containing "active biomass" (biosorption + biodegradation) and "inactivated (autoclaved) biomass" (only biosorption). Abiotic controls under two distinct pH, as utilized for fungi (4.5) and activated sludge (7), were incubated to confirm that pH did not have any influence (like by causing precipitation) on decoloration.

Figure 3 depicts the three dye decoloration performance of fungi and bacteria. Virtually no decoloration was observed in any of the abiotic samples, confirming that the chemical effect of pH can be neglected and decoloration by active/inactivated biomass can be solely attributed to biodegradation and/or biosorption.

Insert Figure 3

Significant decoloration of all the three dyes was exhibited by fungi. Comparison of decoloration of media containing active and inactivated fungi provided evidence that, although biosorption contributed to some extent, the main mechanism of decoloration was biodegradation. For instance, with active fungi, while almost complete decoloration of the copper phthalocyanine dye was observed within 4 days, the corresponding extent of decoloration for inactivated fungi was only 40%. The decoloration of Poly S119 was comparatively slower, 85% decoloration being achieved in 7 days. The decoloration due to biosorption was also lower for this dye (10.4%). However, both the decoloration due to biosorption (3.5%) and total decoloration (46%) of the Reactive Orange 3R was the lowest, suggesting that among the dyes tested, this dye was the most recalcitrant one to fungal degradation.

The decoloration performance of activated sludge was rather poor. A moderate 46% decoloration of copper phthalocyanine dye was achieved. However, as indicated by decoloration of media containing 'active' and 'inactivated' sludge, biosorption was mainly responsible for such decoloration. The decoloration of the rest of the dyes by sludge was very low, only ranging from 1 to 10%, that too mainly due to biosorption. Our observation is in line with the literature reports (e.g., Pagga et al., 1994) that decoloration by aerobic bacteria mainly occurs due to biosorption, and, accordingly, dyes with low biosorption have almost no removal.

Insert Table 3

It is, however, interesting to note that, contrary to poor dye degradation, the TOC removal by bacterial sludge was much better than that by the fungus (Table 3). In case of Poly S119 dye, while approximately 95% TOC removal was achieved in 7 days by the sludge, only 35% was achieved by pure fungus culture. Since in our study contribution of dye to TOC was rather

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low, higher TOC removal by bacterial sludge suggested faster assimilation of relatively easily degradable substances (starch) by bacteria. Coulibaly et al. (2003) also suggested that although white-rot fungus can initiate degradation of wide varieties of recalcitrant compounds, it has limited carbon and nutrient removal capacity as compared to bacteria. Despite differences in dye degradation, no significant difference in TOC removal by fungus in case of different dye solutions was noticed. The same goes for the bacterial sludge. This may again be attributed to the small contribution of dye to TOC.

3.2 Treatment performance of continuous MBR

Preliminary batch tests confirmed the superior dye degradation capacity of fungi. However, the decoloration performance of fungi in the reactors is usually significantly worse than that observed in flask cultures, particularly in the continuous flow reactors (Moreira et al., 1998; Libra et al., 2003; Zhang et al., 1999; Zhang and Yu, 2000). We investigated the TOC and color removal performance of a fungal MBR fed with a synthetic wastewater containing a selected dye— Poly S119.

It is important to point out here that, sterilization of wastewater is extremely expensive and rather impractical in industrial applications. Although the MBR was initially inoculated with pure fungus, in pursuit of development of a practical treatment system, we did not take any specific measures to control bacterial contamination. Accordingly, bacterial contamination did occur. However, a relative abundance of fungi was observed (Table 4) in the MBR during continuous operation.

Insert Table 4

3.2.1 Decoloration

Over 99% removal of Poly S119 dye was consistently achieved in the continuous MBR under an HRT of 1 day (Table 5). In pure culture batch tests, only 85% decoloration of Poly S119 solution was achieved in 7 days. Utilization of different concentrations (1 g/l vs. 5-17.2 g/l) and compositions (pure fungus vs. mixed microbial community dominated by fungi) of the biomass restricts the direct comparison of the rates of removal in batch test and MBR, respectively. However, the excellent removal of Poly S119 dye in the MBR was rather surprising and called for further investigations.

Insert Figure 4
Insert Table 5

Close observation of the quality of the reactor-supernatant (68.3% removal) and the

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membrane-permeate (99.1 % removal) revealed the significant contribution of the membrane to the overall removal performance (Figure 4, Table 5). As is common in case of MBRs, a microfiltration (MF) membrane, which is not expected to remove soluble fractions by its own, was used in this study. Apparently, adsorption of Poly S119 dye on biomass led to its subsequent excellent retention by the cake layer on the MF membrane within the bioreactor. Stable quality of the supernatant as well as the permeate over an operation period of 3 months confirmed that retained dye was subsequently biodegraded. It is important to note here that dye decoloration does not necessarily mean complete degradation. Dye may undergo partial degradation without significant carbon removal, yielding uncolored aromatic substances (Hao et al., 2000). However, in our study significant decrease in the absorbance of the membrane-permeate in the UV range confirmed the subsequent biodegradation of the aromatic group following the breakdown of the color-imparting chromophoric group of the dye (Figure 5).

Insert Figure 5

Interestingly, unlike in pure culture batch test, fungal extracellular enzyme was undetectable in the MBR (data not shown). Since microscopic observation confirmed bacterial contamination in the MBR, it is likely that fungal enzymatic activity or the secreted enzyme itself was significantly destabilized by bacteria. Continuous enzyme washout with effluent may have exacerbated the situation. Libra et al. (2003) demonstrated destabilization of fungal enzyme by a defined bacterial culture in batch test. On the other hand, Zhang and Yu (2000) alluded to detrimental effect of enzyme washout from continuous reactors. Apparently, in our study, the activity of the fungi was just enough to decolorize Poly S119 but not strong enough to show extracellular enzymatic activity. Owing to biosorption and subsequent retention of dye by membrane, the dye degradation rate, although low, was enough to sustain overall excellent decoloration rate. This observation demonstrates the suitability of MBR for practical application of fungal dye degradation.

Results from our study indicate that means to de-couple the dye retention time and HRT of the reactor would allow satisfactory removal even under lower titer of fungal activity in presence of bacterial contamination, while simultaneously avoiding application of excessively long HRT. In the present study, this was achieved by coupling a membrane to the bioreactor. One may, however, wonder about the extent of removal of a recalcitrant dye which shows negligible sorption on biomass (e.g., Remazol Brilliant Orange 3R as used in this study). Investigations in that regard are underway. Preliminary results indicate that addition of adsorbent into MBR may form a sustainable means for removal of chemically diverse dyes.

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3.2.2 TOC removal

Dye bath effluent constitutes only a minor part of TOC in textile wastewater. Unlike the nutrient-deficient hardly biodegradable dye bath effluent, different other streams, namely scouring and desizing effluent, originate from a textile mill. Those streams usually contain high concentrations of relatively easily degradable organics (Robinson et al., 2001). Simultaneous achievement of color and TOC removal is, hence, indispensable.

In addition to excellent decoloration, steadily over 98% TOC removal (Figure 4, Table 5), corresponding to an average TOC concentration of 15 mg/l in the membrane-permeate, was achieved in the MBR. Excellent TOC removal along with decoloration manifested the superiority of the proposed reactor. It is important to point out here that, pure fungi culture can obtain only a moderate TOC removal (Table 3). Fungi can initiate the degradation of recalcitrant compounds (which are not amenable to bacterial degradation); however to complete the TOC removal, bacteria is also required. Hence, although fungal decoloration capacity may be disrupted in presence of bacteria, in order to obtain both color and TOC removal, a reactor design— allowing attainment of fungal dominance to some extent, but also keeping the bacteria in functional state— would be appropriate. Our study shows that MBR can furnish such advantages.

3.2.3 MLSS concentration and reactor stability

A frequently reported problem associated with long-term operation of reactor containing white-rot fungi is the intensive filamentous growth impeding mass transfer and disrupting reactor performance (Zhang et al., 1999). One inherent advantage of MBR is it can sustain much higher MLSS concentration than conventional activated sludge process (Visvanathan et al., 2000). Although MLSS concentrations as high as around 60 g/l have been reported in MBR (Stephenson et al., 2000), in order to limit membrane fouling and reduce cost of aeration, in practice, certain amount of sludge is periodically withdrawn to maintain the MLSS concentration around 10 g/l. In this study, however, to test the stability of treatment performance under high MLSS concentration, sludge was not withdrawn except for MLSS sampling. The initial MLSS concentration of around 5 g/l rose up to 17.2 g/l by day 90 (Figure 6). Since stable color and TOC removals were achieved throughout the operation period (Figure 4), it is evident that, such high MLSS concentration had no detrimental effect on the treatment performance.

Insert Figure 6

3.2.4 Membrane fouling mitigation

Although nowadays MBR has become a reliable alternative to conventional activated sludge

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processes and an option of choice for many domestic and industrial applications, membrane fouling and its consequences in terms of plant maintenance and operating costs limit the widespread application of this technology (Le-Clech et al., 2006). Accordingly, not only achievement of excellent effluent quality but also membrane fouling mitigation is important.

Figure 6 depicts the slight variation of transmembrane pressure (TMP) in the course of operation, during which the MLSS concentration (without any withdrawal of sludge) varied from around 5 to 17.2 g/l. This observation suggested negligible influence of MLSS concentration on hydraulic performance of the membrane in our study. It is evident that under the cleaning strategies, the utilized module effectively resisted fatal fouling despite the high MLSS concentration. Massive accumulation of sludge within and over a membrane module can lead to severe TMP rise and simultaneous acute drop in permeate flux. A previously developed (Hai et al., 2008) spacer-filled module was utilized in this study. Spacer was introduced within the module to minimize the intrusion of sludge by obtaining appropriate compactness under which the fiber arrangement would remain relatively undisturbed, thereby also providing regular backwash channels. The little amount of intruded sludge was backwashed through the bottom end of the module, while the sludge deposited on the surface was effectively cleaned by air-scouring. Accordingly, stable filtration performance could be maintained throughout.

4. Conclusions

This study demonstrates the suitability of membrane bioreactor (MBR) for practical application of fungal dye degradation. Results from our study indicate that means to de-couple the dye retention time and HRT of the reactor would allow satisfactory removal even under lower titer of fungal activity in presence of bacterial contamination, thus simultaneously avoiding application of long HRT. In this study, such de-coupling was achieved by combining a membrane with the bioreactor. Stable decoloration (over 99%) was achieved in the MBR under an HRT of 1 day, and the difference of reactor-supernatant and membrane-permeate quality substantiated the significant contribution of the membrane to the overall dye removal (biosorption, cake layer filtration, biodegradation). This study also revealed that although fungal decoloration capacity may be disrupted in presence of bacteria, in order to obtain both color and TOC removal, a reactor (such as MBR used in this study) which allows attainment of fungal dominance to some extent, but also keeps the bacteria in functional state is required. Over 98% TOC removal was achieved in our study. On the other hand, maintenance of high MLSS concentration (up to an observed value of 17.2 g/l) did not hamper the removal performance in MBR. This is of great importance in terms of plant operation in general and the limitations of conventional fungal reactors in particular.

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Furthermore, membrane fouling—the Achilles' heel of MBR technology— was successfully avoided by utilizing a fouling-resistant compact module. Realization of excellent stable pollutant removal along with alleviation of the membrane-fouling problem manifests the superiority of the proposed system.

Acknowledgments

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List of Tables

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Table 1 Properties of the selected dyes

Dye name	Classification	Color	λ_{max} (nm)	Mol.wt.
Poly S119	Azo (polymeric)	Orange	472	--
Remazol Brilliant Orange 3R	Azo (reactive)	Orange	388	617.54
Copper Phthalocyanine	Metal phthalocyanine (direct)	Blue	630	984.25

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Table 2 Specifications of the membrane module

Item	Unit	Value
Module diameter	cm	4.5
Module height	cm	22
Membrane pore size	μm	0.45
Membrane surface area	m^2	1.07
Average flux*	$m^3/(m^2 .s)$	1.27×10^{-7}

*Operation mode: 5 min on/off

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Table 3 TOC removal in 7 days in batch test

Dye	Media* TOC, mg/l	TOC only by dye,%	Overall TOC removal,%	
			Fungi	Activated sludge
Poly S119	945	5	35	95
Remazol Brilliant Orange 3R	930	4	34	95
Copper Phthalocyanine	930	4	34	95

*Including dye

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Table 4 Relative abundance of fungi/ bacteria in MBR

Stage	Relative abundance*, %	
	Fungi	Bacteria
At the start**	100	0
During continuous operation	65	35

*Size-based approximate fractionation; **Inoculation by pure fungus culture

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Table 5 Average removal performance of the MBR

Average Absorbance		Average dye removal, %		Average TOC removal,%	
Supernatant	Permeate	Supernatant	Permeate	Supernatant	Permeate
1.00	0.03	68.3	99.1	96.8	98.4

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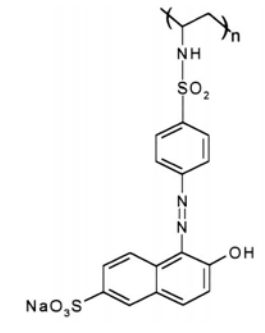
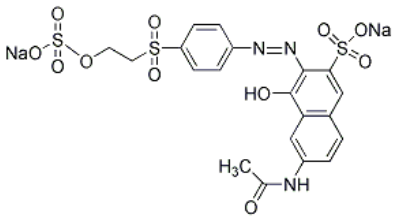
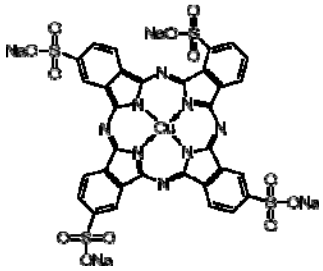
Figure 1 Chemical structures of the utilized dyes



Poly S119

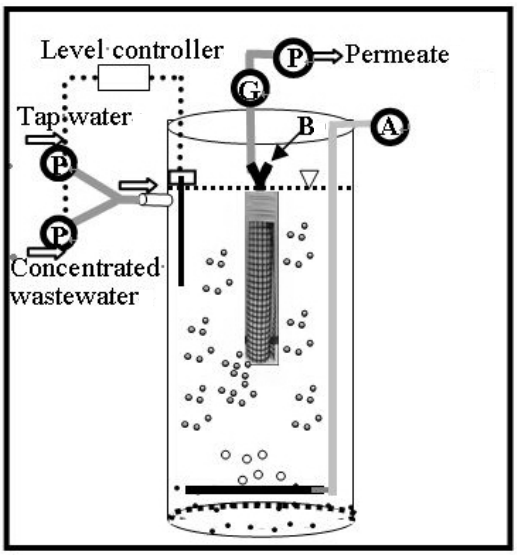


Remazol Brilliant Orange 3R



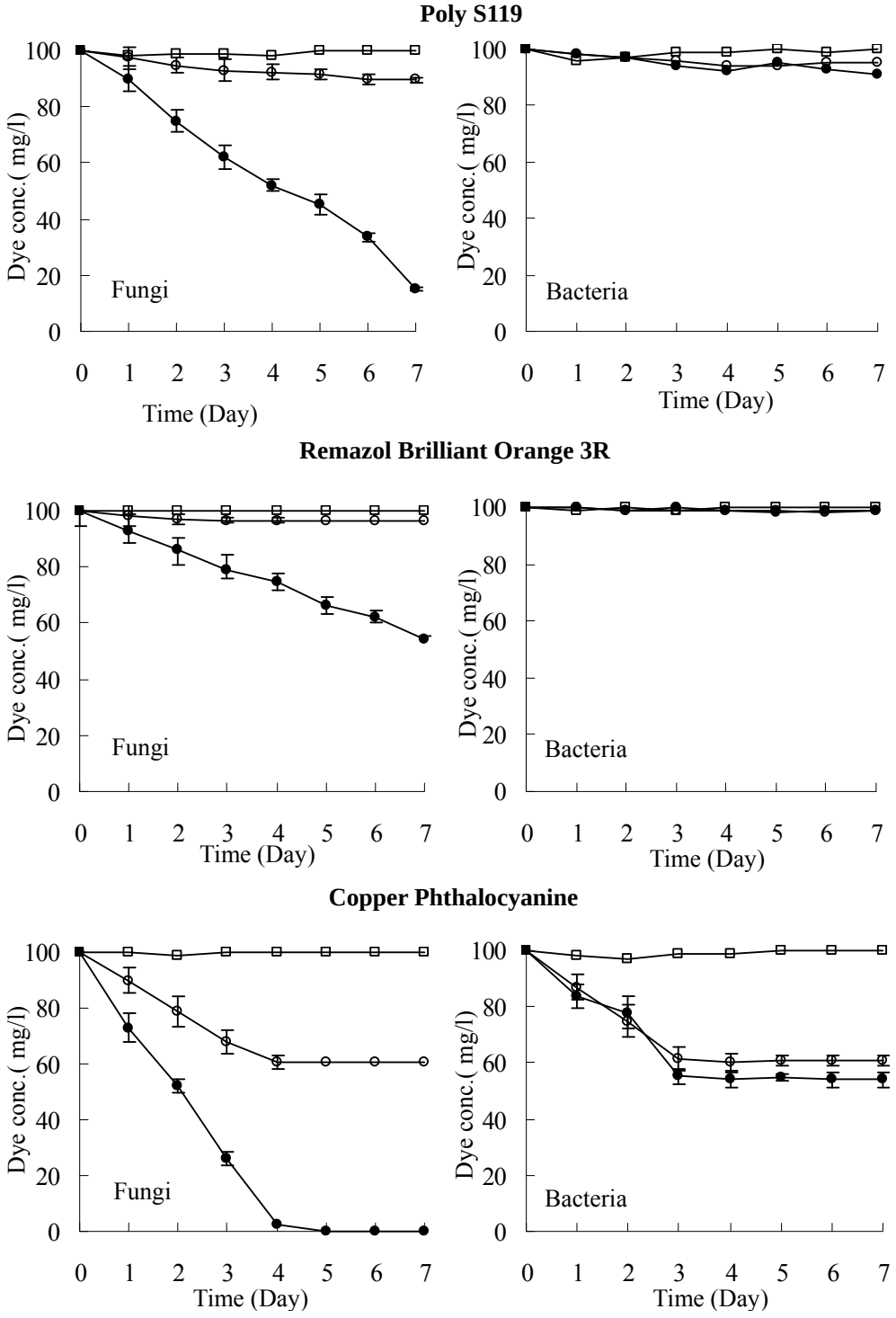
Copper Phthalocyanine

Figure 2 Schematic of laboratory MBR setup



(**A**: Air pump, **B**: Backwash, **G**: Vacuum gauge, **P**: Pump)

Figure 3 Comparison of decoloration by pure fungi and bacteria-dominated activated sludge



Legend: Open circle (inactivated biomass), Solid circle (active biomass), Open square (abiotic)

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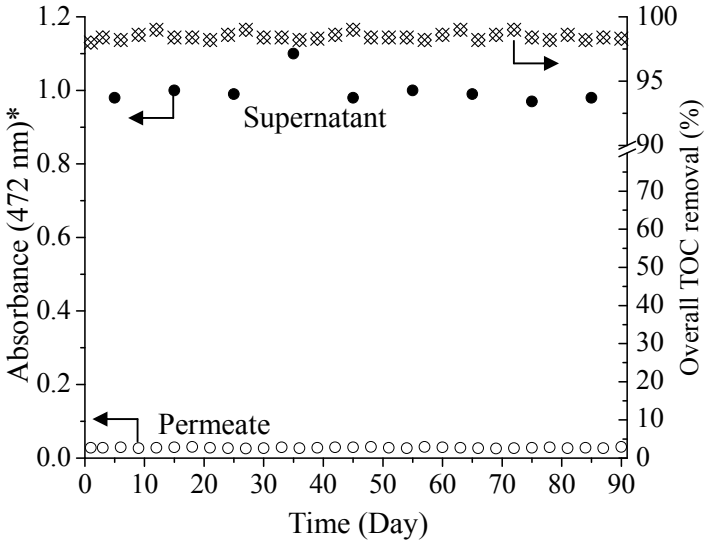
Figure 4 Color and TOC removal in MBR



(*Linear relation between absorbance and dye concentration prevailed in the range shown here.)

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Figure 5 Typical observed change in UV-VIS spectra following MBR treatment

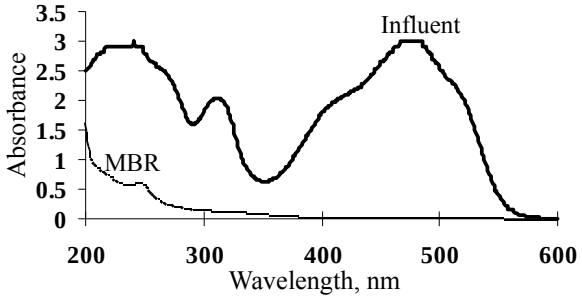

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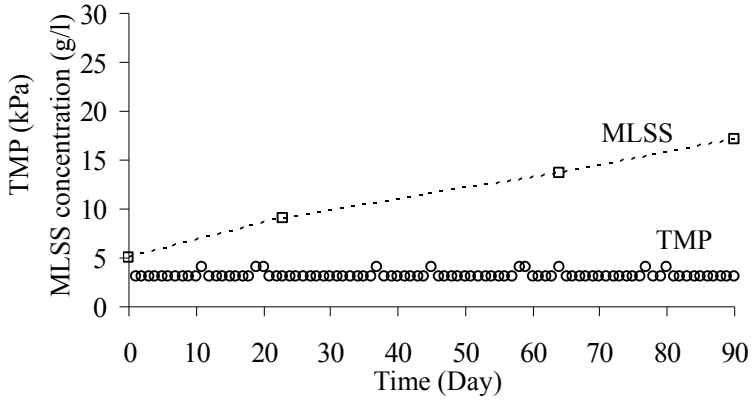
Figure 6 Variation of TMP and MLSS concentration during continuous operation of MBR



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