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Treatment of printing and dyeing wastewater using MBBR followed by membrane separation process

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ABSTRACT

In this study, an assessment of the performance of combined moving bed biofilm reactors and a membrane filtration system (MBBRs, anaerobic–aerobic in series and MF) for the treatment of azo dye reactive brilliant red X-3B-containing wastewater was performed. Each reactor was filled with 35 vol.% of the suspended biofilm carriers for biological treatment. To assess the performance of the hybrid processes, the removal efficiencies of color, COD, and SS were analyzed by experiments. The average removal efficiencies of color, COD, and SS were 90, 85 and 94% (influent color = 400 Pt–Co unit, COD = 500 mg/L, and SS = 310 mg/L), respectively when the hydraulic retention times in the anaerobic and aerobic reactors are 11 and 5 h, respectively. The combined MBBRs and membrane system was highly efficient for the treatment of azo dye–containing wastewater. The experimental results showed that the combined processes are a viable technique for textile wastewater treatment.

Keywords: MBBR; Printing and dyeing wastewater; Membrane filtration; COD; Azo dye

1. Introduction

The textile wastewater discharged from printing and dyeing processes contains a large amount of dyestuff including nitryl, amidocyanogen and heavy metals such as copper, chromium, zinc, arsenic and so on [1]. Thus, textile wastewater is characterized by strong color, large amount of suspended solids, broadly fluctuating pH value varying from 2 to 12, high chemical oxygen demand and biotoxicity and causes coloring of the receiving water environment [2]. Textile wastewater is very difficult to treat satisfactorily due to high variations in their compositions. Strong color is one of the main characteristics of textile wastewater and, if not eliminated, it can cause serious problems to the environment. Azo-reactive dyes are nowadays among the most common classes of dye utilized for cellulosic fibers. During the dyeing process, about 30% of the dye quantity remains in the aqueous phase, mainly in hydrolyzed form, leading to the colorization of the resulting effluent stream [3,4]. Even though azo-reactive dyes are in general less harmful than older types of dyes, many of them have been also found toxic to fish, mammals, as well as, to different kinds of microorganisms.

Various physicochemical (coagulation, membrane filtration [MF], and adsorption), advanced oxidation

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processes (Fenton, ultraviolet, and H_2O_2 oxidation), biological processes (conventional-activated sludge and extended aeration-activated sludge) and usually a combination of processes are applied to treat them to meet regulatory discharge limits [5]. These physicochemical methods are quite expensive have operational problems [6] and generate huge quantities of sludge [7]. Biological methods are generally cheap and simple to apply and have been recently applied to remove organic substances and color of textile wastewater [8]. But the refractory pollutants caused by textile industries cannot easily be degraded by traditional biological process and remain in the effluent. Many attempts have been made to obtain high treatment efficiency of textile wastewater by modifying conventional wastewater treatment process to be combined with additional physical and chemical processes, such as chemical coagulation, electrochemical oxidation and filtration or by applying anaerobic processes, for the treatment of toxic dye materials [9].

Moving bed biofilm reactor (MBBR) as one of the attached-growth biofilm systems was introduced about 15 years old in order to offer the advantages of the former biofilm processes without their limitation, including head loss, clogging, and hydraulic instability [10,11]. MBBR has been efficiently used for the treatment of different municipal and industrial wastewaters during the last decades [12–15]. In the recent years, some reports have been published on successful application of the MBBR process (individually or in combination with other treatment methods) in the biodegradation of some aromatic compounds such as aniline [16], phenol [17], and polycyclic aromatic hydrocarbons and also for the treatment of wastewater containing aerobically recalcitrant compounds [18]. In this reactor, more biomass can be maintained by using various types of carries. 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 very essential phase for biological decolorization. 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 nonspecific enzymes, and these dyes are reduced as electron acceptors. However, intermediate products, such as aromatic amines, can be produced, and these are not further degraded in the anaerobic reactor. The MBBR with anaerobic treatment process followed by and aerobic treatment would be advantageous in treating the intermediate products such as aromatic amines in dyeing wastewater treatment. Membrane separation processes have the potential to produce purified water. Dense membrane processes such as nano- and RO filtration can remove dissolved ions, but their energy consumption is high [19,20]. Microfiltration or ultrafiltration processes, however, seem to be attractive for the separation of particulate contaminants. MBBR-membrane separation process can be operated at relatively low hydraulic retention times (HRTs) or high organic loading rates, since the readily biodegradable matter is removed in the MBBR and particulate matter is separated by the membrane.

The purpose of this present study was to investigate the performance of MBBR followed by membrane separation process for the treatment of wastewater-containing azo dye–reactive brilliant red X-3B.

2. Materials and methods

2.1. Wastewater characteristics

The main objective of the series of experiments carried out with simulated textile wastewater samples was to assess the feasibility of MBBR followed by membrane separation process. Based on desizing, scouring, dyeing, and printing processes in a cotton textile factory, the simulated textile wastewater used in this study exhibited similar composition to real textile wastewater. The characteristics of textile wastewater are shown in Table 1.

2.2. Experimental setup

A schematic of the process configuration is presented in Fig. 1(a). The process consisted of a laboratory-scale MBBR where wastewater-containing reactive brilliant red X-3B was treated and then was fed to the membrane. The MBBR system with an anaerobic–aerobic arrangement was used to treat simulated textile wastewater. The experimental equipment was made of Plexiglas with a length, width, and depth of 70, 25, and 40 cm, respectively. The total volume of the system was 70 L, of which the effective volume was 60 L; the effective volume consisted of the biofilm and membrane zones. MBBR zones consist of

Table 1
Characteristics of textile wastewater

Parameters	Range	Average
pH	10.2–11.8	11.0
COD (mg/L)	400–600	500
BOD (mg/L)	80–120	100
SS (mg/L)	240–380	310
Color, Pt–Co unit	300–500	400


(a) (b)

Fig. 1. Photograph of the reactor.

small cylindrical carriers (Fig. 1(b)) that move freely in the water volume on which a biofilm grows and removes the biodegradable organic matter from the wastewater. The suspended biofilm carriers were obtained from Tongji University (Shanghai, China) and the filling ratio is about 35 vol.%. The carrier elements are made of polyethylene with a density of 0.95–0.98 g/cm^3, which are slightly lower than the density of water. In order to keep the carriers in the reactor, a sieve (with 5 mm opening) is placed at the outlet of the reactor. Textile wastewater was biodegraded by the anaerobic–aerobic biofilm on the moving carriers in the reactor. The filtration unit design in the experiment was based on integrating a flocculation zone below the membrane module where no air scouring was applied, resulting in excellent setting condition for particles in the unit. The sludge collected in the bottom of MF unit was removed once every other day.

2.3. Analytical methods

The performance of the combined system was monitored by analyzing the parameters of COD, SS, color, and pH. All the samples were collected and analyzed during the experiments for almost four months. COD was measured using COD571 (Shanghai Huayan, China), and SS was measured using gravimetric method. The pH value was measured with digital acidimeter PHS-3C (Shanghai REX, China). Decolorization capacity of the processes was determined by absorbance measurements at the maximum visible absorbance wavelength (536 nm). For this purpose, UV–vis absorbance spectra were recorded using an UV–vis spectrophotometer (UV-752, Shanghai, China). Decolorization was determined by the following equation:

$$\text{Decolorization } (\%) = 100(A_0 - A)/A_0$$

where A_0, A expressed absorbency of reactive red dye wastewater in the highest absorption peak place before and after the treatment, respectively.

3. Results and discussion

3.1. Start-up of MBBR

The start-up of MBBR system was extended for four weeks. Due to the low microbial population and nutrient concentration of wastewater, a previous inoculation of MBBR reactor is necessary in order to obtain an adequate biological yield. During the operational period, the sludge from wastewater treatment plant was used as seeding sludge. When activated sludge was used as inoculum, a heterogeneous biofilms on the carriers were obtained, showing several bacterial morphologies and microbial types. It is found that the biomass amount was increased with the time. After four weeks, a thin layer of biofilms could be observed on the inner wall of the carriers. The inside surface area of carrier elements was covered with a brown biofilm. Microscopic tests showed that there were a large numbers of filamentous bacteria in the biofilm adhered to the carrier elements that circulate in the reactor (see Fig. 2), and then, the

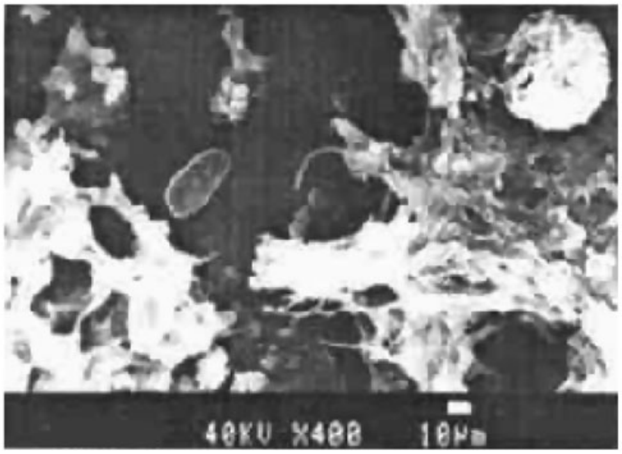

Fig. 2. SEM photograph of the carrier.

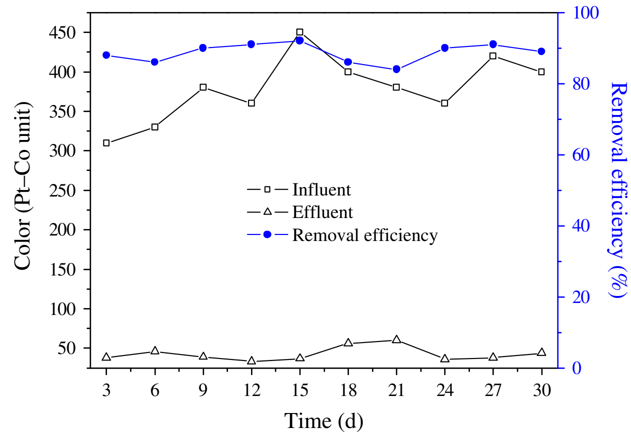

Fig. 3. Removal efficiency of color.

wastewater was changed into continuous flow with initial HRTs at two and a half day. Four weeks later, the COD removal efficiency of the reactor reached nearly 80%, which can be considered as the accomplishment of biofilm cultivation and proliferation.

3.2. Color removal

Azo dyes are resistant to biodegradation under aerobic conditions; however, anaerobic treatment can be implemented satisfactorily. The decolorization of azo dyes invariably begins by reductive cleavage of the azo linkage under anaerobic conditions. This leads to the formation of aromatic amines that are colorless, which may be more toxic than the dye molecules themselves. However, the aromatic amines are easily mineralized under aerobic conditions [21]. In this study, the MBBR system with an anaerobic–aerobic arrangement was used to treat wastewater-containing azo dye. Textile wastewater was feed into the sequenced anaerobic–aerobic MBBR. The carrier packing percentages are 35 vol.%, and the HRTs in the anaerobic and aerobic reactors are 11 and 5 h, respectively. As shown in Fig. 3, the initial high color removal efficiencies observed could be attributed to the adsorption of dye onto the biomass of MBBR. After the 15th day onwards, there was a decrease in color removal efficiencies, and then, it started to increase once again. The nature of color removal pattern showed that dye degradation was occurring in the anaerobic–aerobic MBBR. This can be well supported by comparing the spectra of the influent and the effluent. According to Fig. 3, when color concentration of influent fluctuated from 300 to 500 Pt–Co units, color concentration in the effluent is below 40 Pt–Co units. It can be found that the average color removal efficiency was 90% when the simulated wastewater-containing reactive brilliant red X-3B treated using MBBR was followed by membrane separation process.

3.3. COD removal

In this study, the COD was removed by 20–35% in the anaerobic reactor. Around 60–70% of the COD was degraded in the aerobic reactor. The effect of aerobic phase on COD removal is stronger than that of anaerobic phase. It can be found that COD of textile wastewater decreased under aerobic conditions. Under anaerobic conditions, the color degraded within a certain time, but the COD did not decrease rapidly. As shown in Fig. 4, the average COD removal efficiency was higher than 85%. The system was able to maintain above 85% of COD removal due to the acclimatization of microorganisms to the high concentration of dye. The high removal efficiency could be attributed to both biodegradation and conversion of the aromatic amines and to the most recalcitrant component which will be biodegraded into new components.

3.4. SS removal

Membrane separation is an interesting option since it ensures total retention of suspended matter, significant retention of colloidal matter and presents a barrier of bacteria and most viruses. In this paper, the membrane system was equipped with hollow fiber PVDF membrane. The membrane unit with an asymmetric dense spongy layer and skins formed on both sides of the fiber operated at constant flux. The nominal pore size of the membrane is 0.02 μm. The housing was 1880 mm long with a diameter of

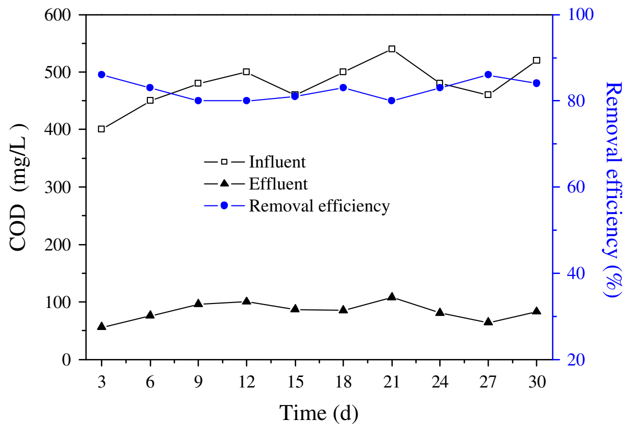

Fig. 4. Removal efficiency of COD.

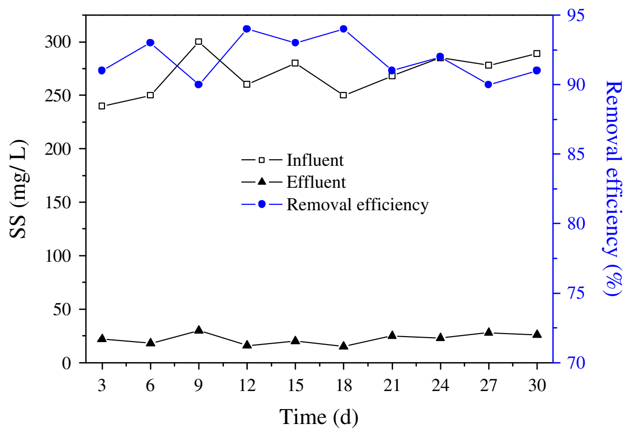

Fig. 5. Removal efficiency of SS.

220 mm. The fiber has an inner diameter of 0.5 mm and an out diameter of 1.25 mm. The total area for filtration was 50 m^2. Fig. 5 shows the SS value of the whole process of influent and effluent. As shown in Fig. 5, the average SS removal efficiency was higher than 94%. The results showed the advantages of the process in SS reduction. The different operating conditions, the pH and effluent flow in the membrane system, did not affect SS reduction because these parameters only modify the biological treatment conditions and not the filtration step.

4. Conclusions

In this study, the effectiveness of laboratory-scale moving-bed biofilm reactors followed by membrane system in printing and dyeing wastewater treatment was investigated. The average removal efficiencies of color, COD, and SS were 90, 85, and 94%, respectively. The color reductions mainly occurred in anaerobic MBBR, while COD and a small amount of color were removed in aerobic MBBR and SS was removed in membrane system. The combined MBBR and membrane system was highly efficient in the treatment of wastewater-containing azo dye-reactive brilliant red X-3B and can satisfy the national guideline of effluent qualities in China. The experimental results showed that the combined process is a viable technique for textile wastewater treatment.

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