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Original Article

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Application of a Moving Bed Biofilm Reactor in Removal of Ciprofloxacin From Real Hospital Effluent: Effect of Operational Conditions

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Abstract

The presence of pharmaceutical wastewater containing antibiotic compound is one of the new problems relating to the environmental pollution. Antibiotic ciprofloxacin (CIP), widely used in medical treatments, can induce antibiotic resistance in low concentrations in the ecosystem and aqueous solutions. In this study, CIP was removed using moving bed biofilm reactor (MBBR) from real hospital-derived wastewater. This study was carried out at Beasat hospital in Hamadan, Iran. CIP (100 mL) was applied in 2 sets of plexiglass tubular columns as MBBR. Microorganisms were grown on the suspended carriers. To achieve this purpose, polyethylene kaldnes (K1) was chosen as reactor bed in 500 m^2/m^3 specific area.

The effect of operating parameters such as mixed liquor suspended solid (MLSS) (100, 1000, 3000 mg/L), hydraulic retention time (HRT) (8, 12, 24 hours), and support media with carrier K1 (30%, 50%, 70%) were evaluated.

According to the results, the yield of CIP removal at 30%, 50%, and 70% of K1, reaction of 24 hours at MLSS 3000 mg/L was obtained 50.5%, 68.9%, and 97.6% respectively. In the same conditions, chemical oxygen demand (COD) removal was achieved 26.78%, 30.49%, and 80.07%, respectively. Results indicated that the MBBR process can be used as an effective approach for removing CIP and COD from hospital effluent. Moreover, these data suggested that the K1 carrier could be useful in terms of mineralization and efficiency. Furthermore, development of biofilm in MBBR was mostly affected by K1.

Keywords: Ciprofloxacin, Antibiotic, Biological process

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1. Introduction

The presence of pharmaceuticals, particularly antibiotics, in aquatic environments has been a growing concern over the past decade (1). These compounds, which are special group of micropollutants, cause environmental contamination through waste disposal of industries as point sources for example and agricultural runoff as non-point sources (2). Recently, effluent from antibiotic manufacturing companies and surface waters downstream of wastewater treatment plants have been reported to contain high levels of antibiotics (3). Residual antibiotics are introduced into sewerage network which finally reach wastewater treatment plants in the forms of original compounds or metabolites. In addition, high concentrations of antibiotics are used, whose disposal increases the attendance in the environment (4). Furthermore, conventional wastewater treatment technologies are not able to effectively remove antibiotics. High levels of CIP have been reported in surface water and effluent (3). The main reason for antibiotic removal is bacterial resistance which is considered a threat to human health. Other concerns about antibiotics are non-target effects of pathogens, restructuring, algae-rich water resources, and intervention in plant photosynthesis caused by abnormalities in the morphology of plants (5).

There are several families of antibiotics, the most important of them are fluorquinolones which have been built in the early 80th century. CIP is a broad-spectrum antimicrobial third-generation fluoroquinolone, and the primary decay product, enrofloxacin, is widely used for the treatment of human infections, and has aquacultural and agricultural applications (6). Although removal of antibiotics from water and wastewater is expensive, their treatment is essential in the industry. So far, numerous methods have been studied for the removal of antibiotic CIP from aquatic environments. Among the most common methods are adsorption, ozonation, and photocatalytic processes (7,8). Considering the costliness and production of intermediate products by the abovementioned methods, biological processes have attracted a great deal of attention thanks to being environment friendly and not needing advanced facilities (9).

Suspended and attached growth processes are methods of activated sludge, however attached- growth processes are effective techniques for wastewater treatment because of the growth of slow-growing microorganisms, which are important for the removal of some micropollutants. On the other hand, attached-growth processes have made different redox conditions within the biofilm. And biomass is not washed out of the reactor through process disturbance (10).

The moving bed bioreactor (MBBR) process is a simple yet effective and compact technology which has been used since 1990. The principle of this reactor is based on the correspondence of active sludge process and fixed bed based on biofilm formation. This reactor, given the type of wastewater, operates aerobically, anaerobically, and anoxically (11). The advantages of MBBR include low head loss, no sludge return, and less cleaning or backwash (5). This process has been successfully used in the treatment of household and sewage wastewater, paper, and cheese industries (12). Dvorak et al investigated the removal of aniline, cyanid and diphenylguanidine from industrial wastewater by MBBR (13). A few studies have been focused on the application of this process in the treatment of hospital wastewater, thus in this research MBBR was run for the removal of CIP from the wastewater sampled from Hamedan's Besat hospital. Moreover, the effects of operating parameters including mixed liquor suspended solid (MLSS), hydraulic retention time (HRT), and support media with carrier (K1) were investigated and the optimization of each process was obtained.

2. Materials and Methods

2.1. Moving bed bioreactor

In this study, a moving bed bioreactor with a volume of 100 L was used in a pilot scale. The reactor was fabricated from plexiglas in a cylindrical form, whose characteristics are provided in Table 1. Airflow was diffused from the bottom of reactor using an air compressor. Hospital raw wastewater was also fed into the reactor from a tank through the peristaltic pump and a valve devised in the bottom of the reactor.

Table 1. Characteristics of the Reactor Used in the Research

Parameter	Value
Material	Plexiglas
Wall thickness (mm)	4
Inner diameter (cm)	30
Total height (cm)	150
Overflow height (cm)	130
Total volume (L)	105
Effective volume (L)	91

2.2. Carrier Bed Characteristics

In this research, carrier K1 was used as the main bed in the reactor. The material of this carrier is polyethylene and polypropylene, whose specific weight is around 0.92-0.96 g/cm^3, usually in the form of wheels with a thickness of 7 mm and diameter of 10 mm. A cross-shaped wall filled them to enhance the strength and the specific area. The specific area of carrier K1 for biofilm growth at 60% of filling was 500 m^2/m^3.

2.3. Experimental Procedure

First, the designed reactor was filled with 30% of carrier K1. Next, one-third of the reactor volume was filled with sludge returning from a secondary sedimentation bath in the hospital's treatment plant. The rest of the reactor volume was filled with raw wastewater so that the reactor was operationalized in a batch way. Thereafter, using an air pump, the aeration rate was adjusted in a way that the dissolved oxygen was supplied by around 2-3.5 mg/L. for operationalization of the MBBR system in winter, and for keeping the optimal temperature for stable growth of biofilm, an automatic aquarium heater was employed and the temperature was adjusted between 20°C and 25°C. Some characteristics of the hospital-derived wastewaters (HDWs) are presented in Table 2.

To accelerate and help biofilm growth, dry milk (Nan Co. Nestle, Iran), containing nutrients and trace elements (Fe, P) required for the growth of microorganisms, was used with a ratio of 33 mg/L for 20 days in 5 occasions. After 4 weeks, a sensible growth of biofilm was observed on the carrier K1. Following this stage, the reactor was operationalized continuously in order to evaluate the effect of different operating conditions on the efficiency of the reactor. In this mode of operation, the rate of the flow fed into the reactor was adjusted between 0.001 and 0.003 L/s. Sampling was carried out at regular intervals and then the samples were centrifuged (5000 rpm for 2 minutes) and analyzed by spectrophotometer at the wavelength of 275 nm (14). The efficiency of COD

Table 2. Characteristics of HDW used in MBBR

Parameter	Value
pH	7.3-8.5
BOD (mg/L)	400-500
COD (mg/L)	750-850
Phosphorus (mg/L)	7.23
Nitrogen (mg/L)	36
TSS (mg/L)	300-400

Abbreviations: TSS, Total suspended solids; COD, chemical oxygen demand; BOD, biochemical oxygen demand; HDW, hospital-derived wastewater; MBBR, moving bed bioreactor.

removal was calculated as follows (15).

$$COD\ removal\ (\%) = \frac{C_0 - Ce}{C_0} \times 100 \qquad (1)$$

Where C_0 and C_e are initial and final concentrations of COD, respectively.

3. Results and Discussion

3.1. Effect of 30% Carrier K1 on Removal of Antibiotic and COD

In order to examine the role of biomass attached to the carrier K1, the biofilm carriers were tested with different fillings. Considering the fact that the volume occupied by the carrier's component does not usually exceed 70% of the total volume of the tank, thus in this study 3 volumes of fillings including 30%, 50%, and 70% were considered by carrier K1. Moreover, different concentrations of MLSS (below 100, 1000, and 3000 mg/L) in the MBBR were selected. The mineralization and biodegradability of CIP were determined by the level of COD removed.

In the first step, 30% of the bed was filled with K1, and the microorganism concentrations below 100, 1000, and 3000 mg/L were studied at residence times of 8, 12, and 24 hours (Figs. 1 and 2). The efficiency of antibiotic removal within 8 hours was 37.7%, 46.4%, and 50.5% respectively, with COD removal percentages of 12.1%, 18.64%, and 26.78%.

Accumulation of the bed is a factor that results in flexibility in the specific space of the biofilm and modification of the reactor's capacity with a certain volume. It is also possible to determine the accumulation of carriers individually for different parameters in MBBR. Carriers in moving bed reactors such as MBBR system increased the population of microorganisms for removal of organic material in wastewater; so the growth rate of biofilm in the reactor can be accelerated. On the other hand, increasing the packing ratio increases the surface for growth of biofilm in tank aeration (16). Accordingly, the average removal efficiency at filling degree of 30% to 70% increases from 50.5% to 76% for CIP and 26.78% to 38.4% for COD. This is in accordance with the finding of Zinatizadeh et al who investigated the removal of carbon and nitrogen from wastewater (10).

3.2. Effect of 50% Carrier K1 in Removal of Antibiotic and COD

As can be seen in Figs. 3 and 4, the efficiency of antibiotic removal in 50% K1 was 48.6%, 56.6%, and 68.9% and COD removal efficiency was 21.01%, 27%, and 30.49%. Increasing the carrier percentage led to the increase in removal efficiency. In this regard, K1 percentage played a main role in this study. MLSS concentration in MBBR process showed the treatment capacity of pollutant. The average removal efficiencies for CIP and BOD increased with the increase of MLSS concentration. In fact, one of the most striking differences between the attached and suspended growth systems is caused by MLSS. Due to the preservation and development of the concentration of microorganisms, biofilm growth was intensified. Therefore it is expected that within longer residence times and biofilm growth increases, the removal efficiency is approved, and hence it can be inferred that MLSS is mainly responsible for the better efficiency of the MBBR system. This result is in line with the finding of Martín-Pascual et

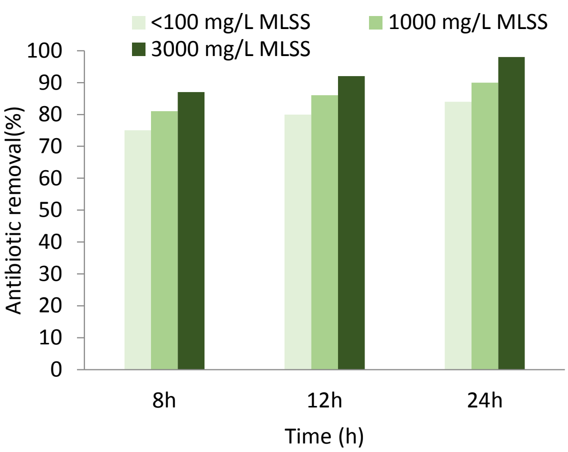

Fig. 1. Effect of 30% Carrier K1 in Removal of Antibiotic.

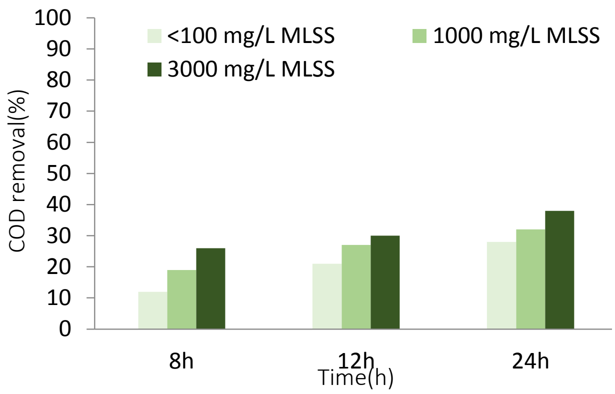

Fig. 2. Effect of 30% Carrier K1 in Removal of COD.

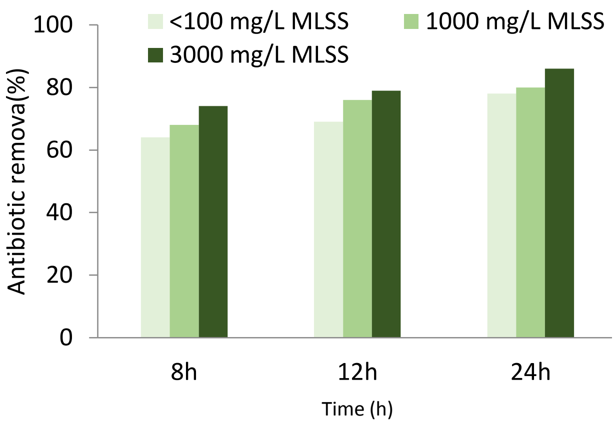

Fig. 3. Effect of 50% Carrier K1 in Removal of Antibiotic.

al who reported that increasing the MLSS concentration in MBBR increased the removal yield of COD and BOD in the treatment of urban wastewater (17).

3.3. Effect of 70% Carrier K1 in Removal of Antibiotic and COD

Finally, according to Figs. 5 and 6 in 70% K1, efficiency for removal of the antibiotic was obtained as 60.2%, 69.5%, and 76% while COD removal efficiency was reported to be 28%, 32.67%, and 38.4%. Regarding these results, the highest efficiency for antibiotic removal obtained at filling percentage of 70% of K1 was 76%. However, within the same time, the rate of the mineralization of this contaminant was reported to be 38.4%.

Based on the obtained results, with the increase in the filling level and prolongation of residence time, the greatest efficiency was obtained to be 86% when the concentration of microorganisms in the system was 3000 mg. Further, the antibiotic removal efficiency, when the bed was filled with 70% of K1, had an ascending trend to such a level that within 24 hours when MLSS concentration was 3000 mg/L, it reached 97.6% and the COD removal efficiency grew to 80.07%. To ensure the growth of microorganisms, they should remain in the system for sufficient time to be able to proliferate. Longer hydraulic residence times influence microbial growth.

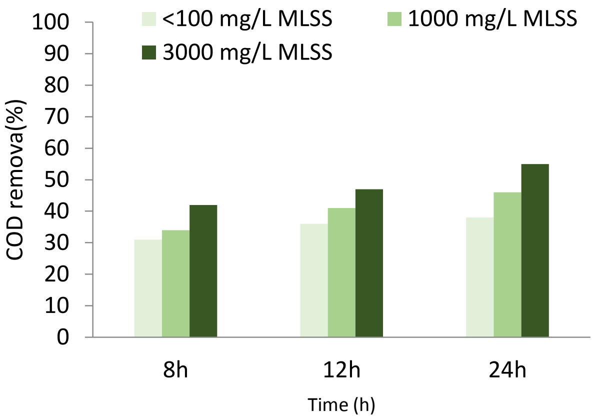

Fig. 4. Effect of 50% Carrier K1 in Removal of COD.

Moreover, elevation of efficiency suggests the propensity of microorganisms present in the living layer of organic compounds and antibiotics. Biological processes can be economic if the contaminant removal efficiency were in high level in low time (18). These results showed the higher efficiency of MBBR at 24-hour reaction time. In this study reaction time played an important role in CIP removal. It can be because of the operation of MBBR set at 8-hour. The biofilm on the packing media was formed with low thickness and the carriers were fluidized on the surface. The increasing effect of HRT on removal efficiency was obtained at 24 hours due to a decrease in dissolved oxygen and the ratio of food to microorganisms (16). A previous study showed that HRT did not have an important parameter in biofilm reactor but another study illustrated that HRT played an effective role in the biological system (10,19). Shokoohi et al also investigated the removal of BOD and COD from hospital wastewater by MBBR. They indicated that under convenient operating conditions of the studied variables (packing rate 70%, HRT 24 hours, and MLSS 3000 mg/L), the removal efficiencies for BOD and COD were 97.8% and 95.6%, respectively (20).

In this study, antibiotic removal was higher than COD removal. Correspondingly, with a large variation in COD removal from the system, the biodegradability of CIP was occurred. The results of our study compared to those of other studies reported the maximum COD removal as 80.07% in 24 hours. Due to the fact that during antibiotic removal processes, various organic intermediates were produced, they needed longer time for degradation by MBBR (21). Therefore, produced intermediates can reduce the MBBR efficiency in terms of COD removal. Moreover, the same finding can be seen in some studies on dye or bisphenol removal (22,23). On the contrary, Jafari et al compared anaerobic fluidized bed reactor (AFBR)/MBBR. They concluded that anaerobic reactor played a most important role in the removal of COD and MBBR was actually needed to polish the anaerobically-treated wastewater and ensure the supply of high-quality final effluent. MBBR system had low efficiency in COD removal (24).

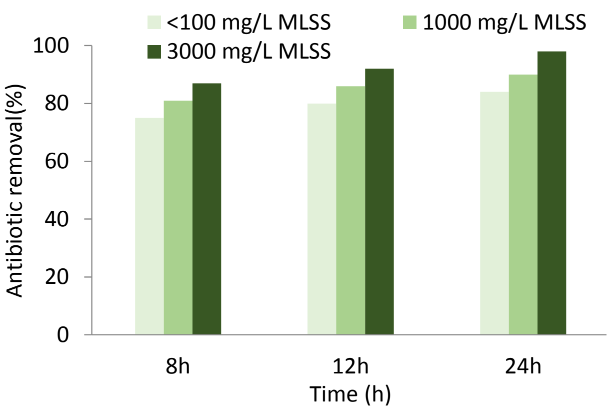

Fig. 5. Effect of 70% Carrier K1 in Removal of Antibiotic.

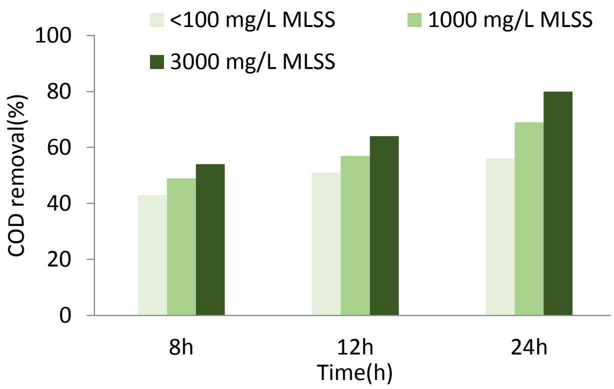

Fig. 6. Effect of 70% Carrier K1 in Removal of COD.

4. Conclusion

In the present study, the performance of MBBR on CIP removal from real hospital effluent was studied. Moreover, the effects of some operational parameters such as MLSS, HRT, and support media with carrier were investigated. Obtained results showed that the maximum efficiency of CIP and COD removal was 97.6% and 80.07% respectively under 70% of K1, and MLSS concentration of 3000 mg/L after 24 hours. Hospital wastewater contains many hazardous materials that separate it from other wastewater. Application of MBBR process has been successful in CIP and COD removal and therefore this method can estimate the standard level.

Conflict of Interest Disclosures

The authors declare that they have no conflict of interests.

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