A Study on Membrane Bioreactor for Water Reuse from the Effluent of Industrial Town Wastewater Treatment Plant

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ABSTRACT

Background: Considering the toxic effects of heavy metals and microbial pathogens in industrial wastewaters, it is necessary to treat metal and microbial contaminated wastewater prior to disposal in the environment. The purpose of this study is to assess the removal of heavy metals pollution and microbial contamination from a mixture of municipal and industrial wastewater using membrane bioreactor.

Methods: A pilot study with a continuous stream was conducted using a 32-L-activated sludge with a flat sheet membrane. Actual wastewater from industrial wastewater treatment plant was used in this study. Membrane bioreactor was operated with a constant flow rate of 4 L/hr and chemical oxygen demand, suspended solids concentration, six heavy metals concentration, and total coliform amounts were recorded during the operation.

Results: High COD, suspended solids, heavy metals, and microbial contamination removal was measured during the experiment. The average removal percentages obtained by the MBR system were 81% for Al, 53% for Fe, 94% for Pb, 91% for Cu, 59% for Ni, and 49% for Cr which indicated the presence of Cu, Ni, and Cr in both soluble and particle forms in mixed liquor while Al, Fe, and Pb were mainly in particulate form. Also, coliforms in the majority of the samples were <140 MPN/100mL that showed that more than 99.9% of total coliform was removed in MBR effluent.

Conclusion: The Membrane Biological Reactor (MBR) showed a good performance to remove heavy metals and microbial matters as well as COD and suspended solids. The effluent quality was suitable for reusing purposes.

Keywords: Heavy Metals, Industrial Wastewater, Membrane Bioreactor, Microbial Matters.

INTRODUCTION

Heavy metals and microbial contaminants have a particular significance in eco-toxicology. Due to the discharge of large amounts of metal-contaminated wastewater, industries bearing heavy metals, such as Cd, Cr, Cu, Ni, As, Pb, and Zn, are the most hazardous among the chemical-intensive industries. Because of their high solubility in aquatic environments, heavy metals can be absorbed by living organisms. Once they enter the food chain, large concentrations of heavy metals may accumulate in the human body. If these metals are ingested beyond the permitted concentration, they can cause serious health disorders such as cancer, organ damage, nervous system damage, etc. [1-5]. The new challenge in wastewater treatment technologies is the removal of these micro pollutants to standard levels.

Membrane filtration is one of the best available techniques in this field. Many studies report the application of this technology to water purification [6], treatment of pesticide industry effluents [7], endocrine disrupting compounds and pharmaceuticals products [8], virus removal [9-11], sterilization [12], organic compounds and microorganisms removal [13-15], and landfill leachate [16-18].

There are different types of membrane filtration, such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO), which can also be classified based on membrane types or based on membrane configurations and modules. In water and wastewater treatment, microfilter and ultrafilter can integrated directly with the activated sludge process to form a technology called membrane bioreactor technology (MBR). There are two configurations for MBRs which are in-series and submerged MBRs [19].
bioreactors have lower power requirements than external MBR configurations [20,21].

Due to the shortage of water resources in the Shokouhieh Industrial Town (located in Qom province, Iran), reclamation and reuse of industrial wastewater treatment plant effluent for usage in some industrial plants was put on the agenda. Effluents of this wastewater treatment plant (WWTP) were not adequately treated by biological treatment and there were heavy metals and biodegradable organic matters in effluent. This research has focused on the evaluation of the pilot scale operation and monitoring of an MBR system to advance the treatment of an industrial wastewater and remove heavy metals and microbial contaminants in order to produce permeate water with high quality. The removal of certain pollution parameters, such as chemical oxygen demand (COD), suspended solids (SS), total coliform (TC), and heavy metals (Al, Fe, Pb, Cu, Ni, and Cr), were monitored.

MATERIALS AND METHODS

Wastewater Source

The actual wastewater used in this study was taken from a wastewater treatment plant in Shokouhieh, Qom, Iran. This plant receives and treats a mixture of municipal and industrial wastewater from different factories such as welding, dairy, beverage, metal finishing, and so forth. The treatment system in this wastewater treatment plant consists of screens, equalization tank, anaerobic reactor, aeration aerobic tank, sedimentation, sand filter, and a disinfection system. Due to poor design, the existing treatment system is not effective in removing all heavy metals and the organic load of influent wastewater. Hence, there is a significant amount of biodegradable organic matters and heavy metal contaminants in effluent. The wastewater samples as MBR feed wastewater were collected from the outlet of sand filters in plastic containers and delivered to the laboratory where the pilot is operated there. The typical physicochemical characteristics of the wastewater are presented in Table 1.

Table 1. Characteristics of feed wastewater.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td></td>
<td>7.3 ±0.62</td>
</tr>
<tr>
<td>SS</td>
<td>mg/L</td>
<td>223 ± 32</td>
</tr>
<tr>
<td>COD</td>
<td>mg/L</td>
<td>250 ± 64</td>
</tr>
<tr>
<td>Al</td>
<td>µg/L</td>
<td>250 ±70</td>
</tr>
<tr>
<td>Fe</td>
<td>µg/L</td>
<td>180 ±80</td>
</tr>
<tr>
<td>Pb</td>
<td>µg/L</td>
<td>340 ±190</td>
</tr>
<tr>
<td>Cu</td>
<td>µg/L</td>
<td>610 ±170</td>
</tr>
<tr>
<td>Ni</td>
<td>µg/L</td>
<td>160 ± 90</td>
</tr>
<tr>
<td>Cr</td>
<td>µg/L</td>
<td>225 ±105</td>
</tr>
</tbody>
</table>

MBR Pilot Unit

Continuous operation of a pilot scale ultrafiltration membrane bioreactor system was carried out in this study. Schematic process flow diagram of the pilot set-up with a picture of the system in operation is shown in Figure 1.

The bioreactor was made of Plexiglass with a total volume of 32 liters. A flat sheet membrane ultrafilter was placed in the center of the bioreactor. The membrane specifications are summarized in Table 2.

Figure 1. Schematic process flow diagram (a) and picture of MBR module in operation (b).
Table 2. Specification of membrane.

<table>
<thead>
<tr>
<th>Value</th>
<th>Unit</th>
<th>Process parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flat sheet</td>
<td></td>
<td>Membrane configuration</td>
</tr>
<tr>
<td>150</td>
<td>kDalton</td>
<td>Cut off</td>
</tr>
<tr>
<td>0.4</td>
<td>µm</td>
<td>Pore size</td>
</tr>
<tr>
<td>240×200</td>
<td>mm</td>
<td>Dimensions (Width × Height)</td>
</tr>
<tr>
<td>0.048</td>
<td>m²</td>
<td>Effective surface area</td>
</tr>
<tr>
<td>EPS</td>
<td>-</td>
<td>Material</td>
</tr>
<tr>
<td>Neutral</td>
<td>-</td>
<td>Membrane charge</td>
</tr>
<tr>
<td>4-11</td>
<td>-</td>
<td>pH resistance range</td>
</tr>
</tbody>
</table>

The membrane operated at a constant flow rate of 4 L/hr using a peristaltic pump. An air blower was used to provide required sufficient air during the operation of the MBR. Air was introduced via perforated plastic tube air diffusers which were located at the bottom of the reactor to produce fine and coarse bubbles for supplying dissolved oxygen required for the biological process in the reactor and reducing fouling on the membrane, respectively. Also, the pilot was equipped with control instruments for measuring temperature, dissolved oxygen (DO), pH, and wastewater level.

**Operating Conditions**

Membrane bioreactor was operated continuously, corresponding to an 8-hour hydraulic retention time (HRT) and the duration of operation after start up stage was 30 days. Prior to use, the membrane was washed with tap water until a steady pure water permeate flux was obtained. The MLSS temperature in the bioreactor was kept constant at 22–27°C with a heat exchanger. Permeate flux was set to approximately 8.3 lm²hr⁻¹ using a peristaltic pump and transmembrane pressure (TMP) was continuously recorded using an analogue pressure gage. Chemical cleaning of the membrane module was not carried out during the operation. During start up stage no biomass was initially removed from the reactor to allow the biomass concentration build up in the system to about 2000 mg/L. After that, daily withdrawal of mixed liquor was conducted from the reactor in order to maintain the predetermined SRT (25 day) and to control an excessive increase of organic matter and solid concentrations in the bioreactor.

**Analytical Method**

Laboratory analyses were conducted to determine the characteristics of influent wastewater to pilot, activated sludge, and MBR permeate. For this, suspended solids (SS), chemical oxygen demand (COD), and concentration of six heavy metals (Al, Fe, Pb, Cu, Ni and Cr) were analyzed. Most analytical techniques used in this study followed the standard methods described by APHA (1998). All data presented in this study was averaged by at least 2 experiment results in each process. MLSS was measured by a Whatman glass microfiber filter using APHA 2540E standard method. The COD content of the samples was measured using Hach COD reactor. To determine metals concentrations, the samples were prepared according to standard methods [15] and then Al, Fe, Pb, Cu, Ni, and Cr values were measured using an atomic absorption spectrometer.

Determination of total coliform was carried out for MBR feed and permeate samples using standard method 9222 B procedure.

The temperature of the MBR pilot (inlet and outlet of the pilot and MLSS temperature in the bioreactor) was monitored using a digital temperature probe (JENWAY, England). These data were verified periodically using an alcohol thermometer. Moreover, MBR influent and effluent pH values were determined using a portable pH meter (JENWAY-370, England) and dissolved oxygen (DO) meter (JENWAY-970, England) was used to determine DO level in the reactor.

**RESULTS**

Before starting the experiments, at startup phase, MBR module was operated for more than five weeks and the stable phase
was obtained. At times, high values of the MLSS concentration were measured and MLSS concentration increased up to a value of around 2000 mg/L. After that, sludge removal was initiated to maintain MLSS concentration constant in the reactor. During the 30 days of the operation of the reactor, MBR performance based on influent and effluent quality and removal percentage data of SS, COD, heavy metals, and TC showed that the system produced permeate water with excellent quality. The percentage removal (R) of each metal was calculated using the following equation:

\[ \text{Removal Percentage} = \frac{C_{in} - C_{out}}{C_{in}} \times 100 \]

Where \( C_{in} \) is the influent concentration and \( C_{out} \) is the permeate concentration of the metal ions.

Figure 2 shows the concentration of SS for inlet and outlet of MBR system and biosolids concentration (MLSS) versus the days of operation. Inlet SS concentration ranged from 179 to 243 mg/L. During this study, it was detected that MLSS of the pilot was within the range of 1600–2300 mg/L. Because of the extended order of magnitudes of the concentration values, the concentration measurements were plotted on a logarithmic scale.

Figure 2. Concentration of SS in the MLSS, inlet and the outlet versus the time of operation.

As shown in Figure 2, excellent solids separation was achieved by the UF membrane. Removal of SS reached greater than 98% resulting in the MBR permeate with SS levels below 3 mg/L.

Figure 3 shows the removal efficiencies of COD for influent and effluent of MBR reactor. COD is a measurement of the oxygen equivalent of the organic material in wastewater that can be oxidized chemically. COD is very important in wastewater treatment because it is an appropriate index representing the organic contaminants in wastewater. Thus, in this study, COD was selected as the organic pollution indicator.

It can be seen that the inlet COD varied from 187 to 314 with the average COD concentration of the influent equal to 222 mg/L whereas COD concentration in effluent varied between 41 and 51 and the average elimination rate was higher than 75%. This indicated that MBR system produced good removal of organic constituents and it was capable of achieving a high removal of COD and organic load could decrease effectively. Some studies reported more than 90% of COD removal which is higher than the results of this study [22-23]. Lower COD removal in this study may relate to less organic material concentration in the bioreactor.

Figure 4 depicts heavy metals concentrations in inlet, MLSS, and outlet of the MBR during the operation time. As Figure 4 illustrates, during the experimental phase, the MBR was able to reduce Al, Fe, Pb, Cu, Ni, and Cr from 250±70 µg/L, 180±80 µg/L, 340±190 µg/L, 610±170 µg/L, 160±90 µg/L, and 220±100 µg/L in feed stream to 45±15µg/L, 85±25 µg/L, 20±10 µg/L, 50±20 µg/L, 60±30 µg/L, and 110±40 µg/L, respectively. This means that 81%, 53%, 94%, 91%, 59%, and 49% of Al, Fe, Pb, Cu, Ni, and Cr were removed, respectively.

In addition to heavy metals and suspended solids, the removal of microorganisms was evaluated. In water and
wastewater treatment systems, total and fecal coliforms were used as indicator organisms. They did not provide direct measurement of pathogenic (disease causing) organisms, but they indicated that pathogenic organisms associated with feces might be present. In other word, if large numbers of fecal coliforms are found in water or wastewater, it is possible that pathogenic organisms, such as Giardia or Cryptosporidium, may also be present [22].

The results of total coliform analyses done on the feed wastewater and MBR permeate are shown in Figure 5. As the results show, total coliform values in the MBR effluent samples ranged from 75 to 140 MPN/100 mL, giving an overall log removal of >4 log for the total coliforms. As Figure 5 shows, there is a trend of decreased permeate coliform with an increasing time of operation. This is expected because as the membranes become clogged, the pore size decreases which results in removal of microorganisms and other particles which could normally pass through the membrane. These data show a high performance of MBR for microorganism reduction.

![Figure 4. Heavy metals concentrations in inlet, outlet, MLSS and their removal percentage.](image)

![Figure 5. Total coliform removal by MBR during operation.](image)
DISCUSSION

The high percentage removal of suspended solids by the MBR indicates that the membrane was in a good condition. MBR suspended solids removal effectiveness as a result of the fact that separation of biosolids by membranes is independent of the bio solid flocculation and solid reduction in permeate water depends on the pore size and the integrity of the membrane.

The high COD reduction implies that a good biodegradable and non-biodegradable COD reduction was achieved through membrane filtration. Most non-biodegradable matters were ultimately removed through sludge wasting. Only a small fraction of non-biodegradable substances passed through the membrane. Similar results were reported by Jianguo (2004) [23]. The effluent COD consists of principally aquatic humic substances, which are naturally occurring compounds. They are hard to biodegrade aerobically and they are responsible for the yellowish color of treated wastewater effluent. These matters may consist of humin, humic, and fulvic acids [23,24].

In the present study, the MBR achieved a high removal of Al, Pb, and Cu (81%, 94%, and 91%, respectively) which indicates that these matters are mostly in particulate form while other metals exist in both particulate and soluble forms in wastewater. Hence, the soluble parts can pass through the membrane and their concentrations in the effluent are relatively significant. Therefore, removal efficiency for Fe, Ni, and Cr (53%, 59%, and 49%, respectively) is less than that of Al, Pb, and Cu. As some studies have reported, the fluctuation in heavy metal removal efficiencies in MBR pilot is attributed to some factors, such as metal competition, changes in pH and MLSS concentrations, and fluctuations in influent metal concentrations [25].

Results from permeate analysis in this study demonstrate that almost complete removal of coliforms can be achieved by using MBR. This was expected since the size of coliform bacteria is larger than the membrane pore size. However, microorganisms can multiply at all kinds of surfaces in the presence of nutrients. As the results show the average concentration of coliforms observed in permeate water is very low. As similar results reported in previous studies indicate [22,26], occurrence of this amount of coliform microorganism in MBR effluent may be related to the bio-film growth in the feed and permeate lines during operation.

Although the nominal pore size of membrane is 0.4 µm, some pore sizes may be larger than 0.4 µm due to a normal distribution of pore sizes. These larger pore sizes may allow some small coliform to pass through the membrane in the experiment. The larger pores in the membrane gradually drawn and when the biofilm was build up on the membrane surface it might act as a filtration barrier to prevent more small microorganisms form passing through the membrane and thus coliform concentration in permeate gradually reduced.

CONCLUSION

Heavy metals present at very low concentrations are toxic for the environment and the aquatic life as well as human health. Therefore, state-of-the-art technologies are used for removal of these pollutants from the environment. As the application of MBR technology for water and wastewater treatment is rapidly expanding every year, the following conclusions drawn from the present study:

- MBR treatment with biomass concentration (MLSS) 2000 mg/L provided an excellent treatment for industrial wastewater treatment effluent.
- The removal of SS reached 99.99% resulting in a MBR permeates with SS levels below 1 mg/L. This demonstrated excellent solids separation is reachable through the UF membrane.
- MBR showed a good reduction in organic and biodegradable matter. The average COD removal was 75% resulting in an effluent with COD ranging between 41 and 51 mg/L.
- Perfect heavy metals removal was also achieved through the operation; Al, Pb,
and Cu were removed completely, indicating that these two metals existed in particulate form, whereas Fe, Cr and Ni were removed by 53%, 49%, and 59%, respectively.

- MBR showed very high removal of total coliforms.

The MBR effluent with such a high quality can be reused within processes industries such as refineries, petrochemical plants and cleaning, recreational water supplies, or discharged to surface waters.

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