Development of Membrane Bioreactor to Membrane Electro-bioreactor for Advanced Treatment of Wastewater

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ABSTRACT: Limited available water resources have rendered wastewater reuse an important issue to specialists in most developed countries, today. The current study works on membrane filtration for treatment of industrial wastewater. By comparing the two methods of membrane bioreactor (MBR) and hybrid membrane electro bioreactor (MEBR) processes, it finds that earlier fouling in the membrane occurs in the first method than the second one. In the membrane electro-bioreactor, in addition to membrane filtration and activated sludge process, the chemical process of electrical coagulation is performed concurrently, wherein the final product quality is improved and the fouling, reduced. In comparison to membrane bioreactor, this method is capable of removing higher percentage of chemical oxygen demand (COD) as an index of organic matters. Accordingly, it is recommended to use the membrane electro-bioreactor method as an alternative to membrane bioreactor for advanced wastewater treatment.

Keywords: reuse, membrane bioreactor, electrical coagulation, membrane fouling.

INTRODUCTION

Reusing wastewater and contaminated water is a problem that has long been a concern for environmental engineers, who are directly responsible for supplying the water demanded by communities. Today, the ability to recycle the wastewater at urban and industrial wastewater treatment plants offers a suitable source for a variety of uses, such as the agriculture and industry, which is due to the high amount of water provided in this way (Saeedi and Hosseinzadeh 2006). By means of new technologies, polluted water can be easily recycled for agricultural or even household uses. The membrane filters with a nanoscale pore can screen 100% of bacteria, viruses, and even small protein units. A brief study of the different efforts made in the world, like ongoing and future plans for industrial and research centers, indicates that treatment is one of the areas of nanotechnology application in water industry that will greatly reduce the cost of water treatment (Qin et al. 2007, Saeedi and Hosseinzadeh 2006, Shevah 2019, Ueda et al. 1997, Wang et al. 2017, Yerri and Piratla 2019). MBR is a new technology, capable of recycling the wastewater by over 98% for

Kent and Farahbakhsh in a study entitled "Evaluating Reverse Osmosis Pretreatment Options through Low Pressure Membrane" at the University of Gulf, reviewed two options for RO pretreatment: 1) Advanced treatment of wastewater from the secondary pond by the hollow membrane (TMF), and 2) Using the membrane bioreactor (Kent et al. 2011). Their research studied the effect of these two methods on reduction of RO fouling amount. Results indicated that the use of membrane bioreactor (MBR) reduced TOC and COD in treated wastewater by 25%. In addition, the simultaneous use of these two methods in RO pretreatment showed that RO fouling with MBR pretreatment was about half of RO fouling with TMF pretreatment. Also, study of RO fouling showed that RO fouling in both cases was of organic and biological type. Another study by Khalili et al. (2009) classified the effective parameters on bioreactor with a volume of 19 liters into three main categories of membrane characteristics, operational variables, and sludge characteristics. It further investigated the effects of membrane type, aeration intensity, and sludge characteristics on membrane fouling. Results showed that the membrane characteristics were effective on the early stages of fouling; though, their influence on the membrane surface through formation of biofilm was lost. Results of aeration test indicated that there was an optimal point of aeration intensity, which increased on the top and bottom of this membrane fouling point. Study of active sludge's characteristics showed that cell metabolism was the most important factor of biopolymers’ fouling in activated sludge. In a study by Farahbakhsh at Gulf University, the effect of pre-treatment on the efficiency of reverse osmosis was investigated in secondary wastewater treatment (Kent et al. 2012, Murphy et al. 2009). This research investigated RO fouling in two modes: 1) using conventional treatment system, and 2) using MBR system as pretreatment. There, the latter mode displayed better performance of MBR system than the former. Both RO systems were used at a similar discharge rate of 4.9 lit / min and a recovery factor of 26%. According to the results, C-RO was obtained after 23 days and MBR-RO reached the maximum pressure after 43 days, indicating the performance of combined MBR-RO system was 47% better than C-RO. In addition, RO output was evaluated based on TOC, COD, total coliforms, nitrate, nitrite, ammonia nitrogen, electrical conductivity, manganese, calcium, sodium, and ammonium, obtained with high efficiency in both cases. Gholam Reza Nabi and Majid Hosseinzadeh et al. conducted a pilot study at University of Tehran (2015), which aimed at evaluating the membrane bioreactor efficiency in advanced industrial wastewater treatment for reverse osmosis pretreatment (Hosseinzadeh et al. 2015). After performing experiments, they managed to remove 98% of the suspended matter, obtaining 75% of high-quality chemical oxygen demand for feeding reverse osmosis unit. The membrane bioreactor (MBR) is one of the most modern biological wastewater treatment systems that, instead of using sedimentation tanks, employs membrane technology to separate water from sludge (Choi et al. 2016, Falizi et al. 2018, Gehlert et al. 2005, Gong et al. 2015, Gündoğdu et al. 2019a, Gündoğdu et al. 2019b). Regarding the membrane pores’ diameter, the outlet water had a very high quality and could be utilized for various industrial uses and even as RO input to produce distilled water. The high
sensitivity of membranes to various types of organic and non-organic impurities makes it necessary to take extensive measures in these units, with the main problem of this treatment type being the membrane’s early fouling, the cost of washing, and replacement of the membrane, itself. In this method, due to early fouling problems of the membrane, a study was conducted to compare the above method with the membrane electro-bioreactor one (MEBR), for which purpose, quality indicators of TSS, COD, SDI, and TMP were measured.

MATERIALS AND METHODS

In the wastewater treatment plant at Faraman Industrial Town, Kermanshah, the sewage first enters the plant and is directed to the balancing tank, where it goes to primary settling tanks, moving toward the aeration tanks. It then arrives at the secondary settling tanks, enters the logging eventually, and goes out. A significant part of biodegradable compounds is removed from the system in aeration basins by microorganisms, with the other part that decomposes later, entering the next stages. Table 1 shows the average quality of raw and treated wastewater in Faraman wastewater treatment plant.

Table 1. Average quality of raw and treated wastewater in Faraman WWTP

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Value</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>-</td>
<td>7.5</td>
<td>8.1</td>
</tr>
<tr>
<td>COD</td>
<td>mg/L</td>
<td>950</td>
<td>140</td>
</tr>
<tr>
<td>TSS</td>
<td>mg/L</td>
<td>350</td>
<td>35</td>
</tr>
</tbody>
</table>

It is worth noting that the numbers in Table 1, above, are the mean of the samples taken. Because the sewage units' quality varies in different times, the quality of operation will differ, too.

This study tries to determine the effectiveness of hybrid membrane electro-bioreactor process in pretreatment of reverse Osmosis to produce industrial water from industrial sludge, treated in wastewater treatment plant. For so doing, industrial wastewater treatment plant of Faraman, Kermanshah, was selected. Hence, apart from discovering parameters required for MEBR reactor operation, its function was also compared with the common membrane bioreactor (MBR) in terms of output quality parameters and the state of membrane fouling. In order to better compare the processes, two parallel pilot reactors with exactly similar sizes and features were used. Their only difference was the metal electrodes, used in MEBR to operate the electrical coagulation process.

The sewage, collected in Faraman industrial town, enters the wastewater treatment plant and goes through the following stages to be treated and eventually leave the plant. The sewage in wastewater treatment plant of Faraman industrial town in Kermanshah moves through preliminary treatment units, balancing tanks, anaerobic and aerobic units, and then goes into the secondary settling tanks. Afterwards, it passes through logging and is discharged into the river. Because of limited water resources and water demand of industries located in the town, part of the treated wastewater is treated again using MBR processes, the pilot reactors of which are placed to attain acceptable quality, so that it can be used in processing industries of Faraman town again.

Looking at similar previous works and methods used in them, it can be easily seen that pilot research reactors had been placed at the final stage, after the secondary settling in the plants, whereas most degradable matters by micro-organisms are generally used in stages preceding it, more precisely in anaerobic and aerobic one. That is why the amount of degradable components at the time of entering pilot reactors with MLSS was as low as about 2000 mg/L to 3000 mg/L. Therefore, to improve the process, reactors must be placed after the exit point of aerobic and anaerobic units. In this case, the amount of MLSS is expected to increase by 6000 to 17000 mg/L. The indicators studied
in this paper included Chemical Oxygen Demand (COD), Biochemical Oxygen Demand (BOD), Dissolved Oxygen (DO), Mixed Liquor Suspended Solids (MLSS), Silt Density Index (SDI), and Oxygen Uptake Rate (OUR).

To know the membrane structure as well as the details of the cake layer, formed on its surface, which causes the fouling, and to compare the functions of MBR and MEBR, the two reactors are compared in terms of the type of fouling and thickness of the layer formed over their surfaces, as can be observed in Figures 1-4.

![Fig. 1. XRF scanning of the membrane surface in MBR reactor](image1.png)

![Fig. 2. XRF scanning of the membrane cross-section in MBR reactor](image2.png)
Fig. 3. XRF scanning of the membrane surface in MEBR reactor

Fig. 4. XRF scanning of the membrane cross-section in MEBR reactor

Fig. 5. Pilot reactor display while in operation
RESULTS AND DISCUSSION

The output quality of both reactors is examined based on COD and MLSS parameters. The output product of both reactors is transparent due to the membrane process; though, in MEBR it is more transparent than MBR. Nonetheless, the main criterion is the results from the experiments and their comparison. After 10 days, the concentration of suspended matter was 3200 mg/L for MBR and 6600 mg/L for MEBR, leading into the removal of organic matter by microorganisms and COD content, reduced at the outlet. The following Tables and Figs show the reactors' operation during the 48-day period.

Results from examination of the output quality of MBR and MEBR and their comparison.
The amount of MBR's suspended solids (MLSS) varied from 3200 mg/L to 5200 mg/L, and as for MEBR, it ranged between 6600 mg/L and 11000 mg/L, due to the range of many numbers. Logarithmic Figs. will be used to represent the results. Fig. shows the amount of chemical oxygen demand (COD) and Fig. the amount of suspended solids removed (MLSS), measured through a physical process of separation through membranes. After 48 days of both reactors’ operation, COD changes displayed a trend, which can be seen in Figures 8 and 9. The amount of COD of the input wastewater to the reactors varied from 285 mg/L to 560 mg/L, making the mean COD value of the reactor's input, 450 mg/L. After refinement, this value ranged between 54 mg/L and 79 mg/L for MBR, and between 22 mg/L and 31 mg/L for MEBR. Thus, it can be concluded that the removal rate of COD in the output product was 88% and 95%, respectively. Since electrical coagulation process in MEBR takes place relative to MBR reactor, it leads to COD reduction, rendering the output of the first reactor more qualitative than the second one.

Fig. 8. Changes in suspended matter and removal percentage during MBR operation

Fig. 9. Changes in suspended matter and removal percentage during MEBR operation
Over time and following wastewater treatment by means of membranes (membrane filtration), the pores gradually began to clog. The reason behind this phenomenon was that the experiment had been performed by pre-static pumps with a constant flow of 1.5 L/hr. With the flow rate of the membranes remaining constant, the pressure drop increased, because the gradual fouling and subsequent reduction of the membrane pores of the reactors increased the pressure difference in the two ends of the membrane and the permeability pressure of the membrane.

By increasing the duration of the operation along with the membrane's fouling, the range of these changes expanded so that the rate by which the pressure increased was much higher than the time. Under these conditions, the membrane was removed from the reactors and was first washed physically via surface washing with clean water and then chemically. It was then placed inside the reactor once more so that the treatment process would continue. Comparing the two MBR and MEBR reactors, operating with different MLSS, showed that by increasing MLSS, the membrane fouling increased and critical pressure drop occurred faster. Therefore, the time between the membrane washing times became lower. The number of membrane washings in MBR was higher than that of MEBR.
Fig. 12. TMP changes during MBR operation

Fig. 13. Changes in membrane permeation pressure during MEBR operation

Fig. 14. Comparison of changes in membrane permeation pressure during MBR and MEBR operation
This indicates that membrane fouling of MBR occurred faster, because by conducting electrical coagulation process in MEBR and/or releasing metal cations (aluminum) in the reactor, a significant amount of fine particles, more often than not colloids, got removed from the solution and deposited. As a result, fewer particles passed through the pores allowing the membranes to be used for longer periods of time.

**Comparison and examination of the use of MBR and MEBR outlets as pretreatment for reverse osmosis (RO) system**

SDI is one of the main indicators to measure sedimentation and water fouling to enter reverse osmosis modules, where the ratio of sedimentation can be measured based on the amount of sediment, found in the 0.45 Mm membrane at the time of treatment. Typically, in order to reduce sedimentation of water supply and feeding in reverse osmosis units and nano filters, SDI value must fall below 5, within 15 minutes after pretreatment. If SDI indicator of input water into the membrane goes beyond 5, all membranes will be severely fouled and production will be reduced in less than a few weeks. The best SDI for the water entering the reverse osmosis membrane is less than 3. If the indicator is between 3 and 5, then a maximum of 6 months of repeated washing is needed for membranes, which will finally reduce their lifetime.

![Fig. 15. Comparison of SDI indicator after MBR and MEBR outputs at different time intervals](image)

According to the above Fig., three times the SDI value for MBR stood above 3, while for MEBR, it occurred only once. This indicates that the output quality of both reactors was acceptable for reverse osmosis pre-treatment. However, it is worth noting that SDI indicator for MEBR showed a higher quality than MBR under the same conditions.

**CONCLUSION**

As surface and groundwater sources are limited in Iran and water, in general, is an important issue, wastewater recycling by means of new methods is of great importance. In some wastewater treatment plants, there have been some attempts to recycle part of the wastewater through reverse osmosis process so that it can be used in different areas such as municipal landscapes or industrial towns. One of the ways, used as reverse osmosis pretreatment as well as advanced treatment of domestic sewage and industrial wastewater, is membrane bioreactor process (MBR), which instead of gravity separation method
uses the membrane to separate solids from wastewater. Although MBR has resolved many problems related to wastewater treatment, membrane fouling has continued to be the major issue in relation to this method, with several researches conducted in recent years aiming at better development of the method as well as minimizing the problem.

This paper studied the use of membrane bioreactor system for advanced treatment of industrial wastewater, discharged from Faraman wastewater treatment plant to feed the reverse osmosis unit. It investigated MBR output by measuring quality parameters to supply the water, required to enter the reverse osmosis system. Moreover, it dealt with the effect of increased concentration of Mixed Liquor Suspended Solids (MLSS) on membrane fouling as well as on quality parameters of wastewater, discharged from membranes. Filter fouling in membrane bioreactor process is the most important obstacle in the process, which generally occurs due to the presence of colloid particles in wastewater discharged from filters. In the next stage, the effect of this hybrid process on improving the output quality and decreasing the membrane fouling was studied.

After more than 100 days of research to utilize a system as a reverse osmosis pre-treatment better, it can be claimed that according to previous studies, pre-treatment of reverse osmosis decreed making a real and logical comparison between the two MBR and MEBR systems. The wastewater from industrial units of Faraman town, which entered the reactors, first flowed into the treatment plant and after undergoing pre-purification operations, like scavenging, granulation, and entering aerobic unit, passed through aeration ponds, where a significant portion of biodegradable matter was used by microorganisms, thence to be used as feed for reactors. Since the electrolysis factor was of great importance in MEBR method, for the sake of more efficient and accurate operation over a period of about three months, four main options including: 1) pH value, 2) electrode distance (d), 3) value of the voltage (V), and 4) time (t) were controlled in such a way that during treatment in the main reactor, the microorganisms were protected in electrolysis flow and had appropriate pH, logical electrode distance from one another, and correct power voltage supply in a given time period. As such, better wastewater treatment was performed, compared to MBR method, showing firstly reduced amount of COD and secondly significant delay in the time of membranes’ fouling. Together, these results enabled better treatment with lower costs (delay in membrane fouling).

Finally, results from comparison of two pilot reactors of MBR and MEBR can be summarized as follows:

- The average concentration of mixed liquor suspended solids in MBR was 3500 mg/L with its effect on COD removal being 92%, which implies high efficiency of the reactor in complementary wastewater treatment.
- Looking at heavy metals of Al, Fe, Cr, Cu, Pb, Ni, and Zn present in input wastewater and output of the reactor indicated average removal of 63%, 78%, 41%, 83%, 93%, 64%, and 69% for these elements, respectively. High removal percentage of Fe, Cu, and Pb implies that these metals are often present in the reactor in suspended form. Therefore, when they pass through the membrane, they are trapped inside its pores and therefore there would be a low concentration of these metals in reactor outlet. On the contrary, other metals, present in the form of suspension or solution, can pass through the membrane making them less likely to get removed.
- Several experiments with SDI
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indicated that its value was often below 3, proving that the membrane's water output had a very high quality to be used in RO system.

- Electrocoagulation of input wastewater (without using a membrane) led to the following optimum conditions for electrical coagulation in MBR: PH = 7, electrolysis time (t) = 150, electrode distance (d) = 4cm, and electric field = 3V/cm. By performing the present experiment, the optimum electric field was determined in a way not to distort the microorganisms' operation present in activated sludge.

- Assessment and comparison of MBR and MEBR performance with two different MLSS indicated that using MEBR with higher MLSS would improve the output quality, boosting the efficiency of pollutant removal (COD, heavy metals, and suspended matter).

- Comparison of MBR and MEBR reactors' results proved high efficiency of these two processes in removal of organic materials, suspended matter, and heavy metals. The average efficiency of COD and TSS removal in MBR was 80% and 99%, while it was 85% and 99% in MEBR, respectively.

- Concerning heavy metals, removal efficiency of MEBR was higher than MBR in all cases except iron. The average removal of Al, Fe, Cr, Cu, Pb, Ni, and Zn was 63%, 78%, 41%, 83%, 93%, 64%, and 69% in MBR, respectively, whereas in MEBR these figures rose to 60%, 82%, 46%, 92%, 94%, 79%, and 75%. High removal percentage of Fe, Pb, and Cu indicated that these metals were often present in suspension form, while other metals were present in both suspension and solution forms. The solution could pass through the membrane, making it less likely to remove from the bioreactor. Lower removal efficiency of Aluminum in MEBR process resulted from the release of Aluminum ions in electric coagulation process, existing in the solution phase. Passing through membrane pores, its split and removal efficiency declined.

- MBR requires more frequent membrane washing than MEBR, proving that fouling occurred faster in the MBR membrane.

- XRF results showed that MBR sediments mainly included organic materials and Calcium Oxide, while the sediments in MEBR membrane contained Aluminum Oxide as well. Therefore, results indicated that apart from biological fouling, organic materials and colloidal particles were responsible for most of membrane fouling.

- In both MBR and MEBR, the output water to be used in reverse osmosis module, based on SDI index, showed that the quality of water discharged from membrane had been good enough to be used in the given module; however, MEBR had a better and more adequate quality.

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CONFLICT OF INTEREST
The authors declare that there is not any
conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING
No life science threat was practiced in this research.

REFERENCES


