

Original Research Article

Membrane Bioreactor System for Real Wastewater Treatment from a Methanol-to-Propylene Conversion Unit

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ABSTRACT

A membrane bioreactor is actually an advanced system of conventional activated sludge system. The development of MBR systems has solved some of the problems of conventional activated sludge systems that usually exist in the secondary sedimentation unit and during the sludge separation from wastewater. In the secondary sedimentation pond of the conventional activated sludge system, gravity is used to settle the sludge. To set up the pilot membrane bioreactor and easy access to the effluent, the pilot was placed near the methanol to propylene conversion unit located in the center of the special area of Petrochemical Research and Technology Company, and the effluent from tower number 351 of the unit was fed into the unit using a flexible pipe. The pilot feed tank is transferred. The effluent of Tower No. 351 used in this study has a COD of about 20,000, a TDS of about 2,500 ppm, and a turbidity of about 110 NTU, which significantly decreased after membrane treatment. It shows that its majority consists of methanol and a small amount of compounds such as dimethyl ether, acetylene, and methyl ethyl ketone were also observed.

Introduction

Since the last years of the 20th century, lack of water and drought have shown themselves as a widespread crisis. In 2023, about 1.4 billion people in the world will be deprived of access to safe drinking water. World assemblies, especially the United Nations, emphasized the importance of the water crisis and the necessity of its address by holding world conferences and summits. This led to 2005 to 2015 being named Water for Life [1]. The related predictions show

stress, which will have 7 billion people population by 2025, will be the first victims of the water crisis [2]. These predictions have been made based on international indices, among which we can mention the Falcon Mark index and the United Nations index, which respectively per capita water consumption is less than 1000 cubic meters per year and the withdrawal of more than 40% of the renewable water sources have been identified as the water crisis [3]. One of the main factors causing this crisis is the lack of renewable water sources, pollution of water sources caused by industrial,

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urban, and agricultural sewage, increasing water consumption, and security issues. Nowadays, membrane bioreactors and other new membrane technologies have been suggested as a new way to prevent the arbitrary extraction of fresh water, such as running and underground water [4]. Our country, Iran, is located in a dry and semi-arid climate, so it seems that due to the growth of population, industry, and agriculture, Iran will also face the problem of water supply in the future [5]. The conventional activated sludge process generally consists of an aeration pond in which the biological removal of pollution in the wastewater is carried out and a sedimentation pond in which the biomass separation from the treated sewage is carried out by gravity. The main problem in the conventional activated sludge system is related to the stage of separation of biomass from wastewater, and the quality of the process of separating sludge from wastewater is directly related to its sedimentation characteristics, and if the sludge in the aeration pond does not have suitable sedimentation characteristics. The water coming out of the sedimentation pond will not be of good quality, and a large amount of suspended solids will be observed in it, and it will also be difficult to return the sludge from the sedimentation pond to the aeration pond. In the membrane bioreactor system, the role of the sedimentation basin is entrusted to the membrane. The relative density of sludge is usually around 1.02, which is very close to the relative density of water, and weak sedimentation is observed in the secondary sedimentation unit at normal hydraulic retention times of 2 to 3 hours. The problem of sludge settling is mostly related to the size of particles (smaller than 10 micrometers) and bacteria with stringy structures [6]. Lumping or bulking sludge is a common problem in the sedimentation process of conventional activated sludge systems in the conventional activated sludge process. Likewise, membrane modules can be used in two ways to separate sludge from water. In the first case, the membrane module is placed outside the biological reactor chamber, and in the second case, it is placed inside it. The first mode is called the external membrane bioreactor and

the second mode is called the submerged membrane bioreactor [7]. In the external bioreactors, the membrane is placed outside the aeration chamber, and the filtered water flows out from one side of the membrane and the water with the sludge concentration in it is the reason for the exit of some water in filtered form from the membrane, increased, is returned to the aeration chamber. In these bioreactors, the pressure difference and the flow rate on the membrane are two important factors that affect the amount of water coming out of the membrane. These types of systems were used more in the early 1990s, but gradually, due to the lack of use of a pump to return the sludge to the submerged membrane bioreactors and as a result, less energy consumption, this type of bioreactors took their place [8]. They were immersed in membrane bioreactors. However, the use of these bioreactors in special cases, such as wastewaters with very high pollution load, high temperature, very high or low pH, high toxicity, and low filtration capacity, is still of interest and has completely taken their place. The most important difference between submerged and external membrane bioreactors is their energy consumption so that due to the absence of sludge return pump in submerged membrane bioreactors, these types of bioreactors consume less energy [9,10]. The costs of the submerged membrane bioreactor are lower than the external membrane bioreactor in two aspects, one in terms of less energy consumption and the other in terms of the need for washing, because the air used in the submerged membrane bioreactor also plays a role in it is responsible for the oxygen required for the growth and metabolism of microorganisms and also the role of washing the membrane. On the other hand, since the submerged membrane bioreactors operate at a lower flux, a larger surface area of the membrane is needed, which causes the initial cost of constructing the submerged membrane bioreactor units to be higher than the external case.

The characteristics of the pilot used in this study

The pilot designed and built in this study has the following specifications:

- 1- Two-stage process (anoxic and aerobic wastewater treatment)
- 2- The volume of feed to be treated is 2000 liters per day
- 3- The membrane used in the reactor: flat plate
- 4- The type of membrane used in the reactor: Polyvinylidene difluoride
- 5- Equipped with a temperature, pH, dissolved oxygen control system, automatic sludge discharge, and automatic injection of materials required for the biological process.
- 6- The system is portable and has been set up in a shed.

Description

The initial inoculum sludge was prepared from the aeration pond of the LOW TDS unit of FAJR Persian Gulf Energy Petrochemical Company and was poured into the reactor after altering the concentration to 5500 ppm. The inoculation sludge has a volumetric index (SVI) of 130 and an initial concentration of about 2600 ppm. The COD removal efficiency in the pilot is very good and the output COD has always been below 100 mg/L, so it is necessary to have a standard for environmental discharge. The value of the pH parameter inside the bioreactor has been maintained in the neutral range by injecting sodium bicarbonate. Likewise, the nitrogen and phosphorus required by the microorganisms inside the bioreactor have been provided by injecting di-ammonium phosphate chemical fertilizer and phosphate fertilizer, respectively. In this project, due to the limitedness of the actual wastewater, the use of hydraulic retention time (HRT) equal to 18 hours during the operation period of one month was considered. The TDS value of this effluent was 1430 and at the end of one month, it reached 1680, which was due to the injection of nutrients and the source of carbon and phosphorus. An infinite sludge retention time (SRT) was employed in this project by avoiding sludge return to the bioreactor. The only operating variable used in this project is hydraulic retention time. It should be noted that according to the process data, the amount of wastewater produced in the methanol to propylene conversion demo unit is 84 kg per

hour with a COD of about 150 ppm, in the hundred and twenty thousand ton industrial unit, 32110 kg per hour and in a 470 thousand tons industrial unit is 120,000 kilograms per hour, and by treating this amount of wastewater, it is possible to save a lot in the production of water needed for irrigation of green spaces and water for cooling towers. The unique characteristics of the effluent used in this work distinguish it from those studied in previous research, highlighting the innovative nature of this study. Furthermore, similar effluents were not seen at the level of petrochemical companies. The effluent of Tower No. 351 used in this research has a COD of about 20000, TDS of about 2500 ppm, and a turbidity of about 110 NTU. The results of the analysis of the compounds of this effluent show that the majority of it consists of methanol and a small amount of compounds such as dimethyl ether, acetone, and methyl ethyl ketone were also observed. The amount of turbidity in the diluted feed entering the membrane bioreactor system was in the range of 60 NTU, and after purification by the system, the turbidity in the output was always in the range of 1.5 NTU.

The initial inoculum sludge was prepared from the aeration pond of the LOW TDS unit of FAJR Gulf Energy Petrochemical Company (Figure 1). After being concentrated to 5500 ppm, it was poured into the reactor. The volumetric index (SVI) of the inoculation sludge is 130, and its initial concentration is approximately 2600 ppm. The value of the pH parameter inside the bioreactor has been maintained in the neutral range by injecting sodium bicarbonate. In addition, the nitrogen and phosphorus required by the microorganisms inside the bioreactor have been provided by injecting di-ammonium phosphate chemical fertilizer and phosphate fertilizer, respectively.

To provide the necessary elements for the growth of microorganisms, the COD of the feed tank inlet effluent was daily measured, and a nitrogen source and a phosphorus source were added to the system with a COD: N:P ratio of 100:5:1.

Micronutrients were further added to the system according to Table 1. Although the concentration of micronutrients is very low, it

should be noted that their absence can reduce the removal efficiency by 30%. According to the feed COD, the amount of micronutrients (nutrients) required for microorganisms will be different. Therefore, stock solutions with specific concentrations of these nutrients are prepared, whose concentration is calculated based on COD=1000 ppm of the feed, and for

each 1000-liter feed tank of the MBR unit, 20 ml of these nutrient solutions are added to the feed. It is added to provide the nutrients needed by the project. In this way, the concentration of stock solutions and the final concentration of the nutrient feed are calculated with the above explanation for COD=1000 ppm in [Table 1](#).



Figure 1: Sludge transfer from FAJR energy petrochemical company of the Persian Gulf

Table 1: The concentration of nutrient stock solutions used in the project

No.	Nutrient	Stock solution concentration (g/l or ml/l)	Final concentration in the feed (g/l or ml/l)
1	Urea (CH ₄ N ₂ O)	1071.734475	0.02143469
2	H ₃ PO ₄	182.8993563	0.003657987
	CaCO ₃	66.6018038	0.001332036
3	KOH	95.72122062	0.001914424
4	MgSO ₄ .7H ₂ O	202.8641975	0.004057284
5	Fe(NO ₃) ₃ .9H ₂ O	86.82327663	0.001736466
	MnCl ₂ .4H ₂ O	0.468274481	9.36549E-6
6	Zn(NO ₃) ₂ .6H ₂ O	1.81972778	3.63946E-5
	NiCl ₂ .6H ₂ O	2.71367694	5.42735E-5
	CuSO ₄ .5H ₂ O	0.275021243	5.50042E-6
	CoCl ₂ .6H ₂ O	2.018751061	4.0375E-5
	H ₃ BO ₃	2.859851989	5.7197E-5

In this project, the use of hydraulic retention time (HRT) equal to 18 hours was considered during the operation period. The TDS value of this effluent was 1430 and at the end of the one-month period it reached 1680. This increase is due to the injection of nutrients and carbon source and it was phosphorus.

Through this project, there is no return of sludge to the bioreactor, which indicates the infinite SRT. The amount of aeration in this

project has always been such that the amount of dissolved oxygen in the bioreactor is maintained between 2 and 5 ppm. Accordingly, the airline connected to the flowmeter has been used, set to half a cubic meter per hour.

Normally, the amount of NaHCO₃/1700-300 mg per liter of incoming wastewater can provide conditions that prevent acidification of the reactor environment. In this project, 500 mg of sodium bicarbonate per liter of water was

used for this purpose, 1000 mg/liter of diammonium phosphate chemical fertilizer was used as a nitrogen source and 200 mg/liter of phosphate chemical fertilizer was used as a phosphorus source. The material of the membranes used in this work is polyvinylidene di-fluoride in the number of 8 pieces with dimensions of 1 meter by half a meter and thickness of 5 cm. The filtration mechanism is of ultrafiltration type.

Discussion

Laboratory results using real wastewater are compiled in Table 2.

COD test results

The pilot consistently achieved a COD removal efficiency exceeding 99%, indicating compliance with environmental discharge

standards. Initially, COD removal efficiency was approximately 65%. However, microbial acclimation occurred within four days, resulting in a significant improvement in performance. In one month after the adaptation of the sludge, the daily test results (30 days) show that the COD removal efficiency was not less than 85% (Table 2 and Figure 2).

MLSS and MLVSS test results

As indicated in Table 2, the MLSS/MLVSS ratio was in the range of 0.6 to 0.85. This ratio shows the accumulation of inorganic compounds inside the system. An increase in this number indicates that inorganic compounds have accumulated less in the system. The decrease of this number indicates the accumulation of organic compounds inside the system (Figure 3).

Table 2: Laboratory results of the project using real wastewater

HRT (hr)	Day	COD feed (mg/L)	COD permeate (mg/L)	MLSS (mg/L)	MLVSS (mg/L)	MLVSS/MLSS	SVI (mL/g)
18	1	850	77	5710	3450	0.60420	131
18	2	810	72	5430	3505	0.64548	128
18	3	735	63	5620	3200	0.56939	114
18	5	770	59	5380	3705	0.68866	119
18	6	700	54	5050	3510	0.69504	108
18	8	910	94	4840	3620	0.74793	104
18	9	925	91	4970	3570	0.71830	109
18	11	890	75	5030	3510	0.69781	100
18	12	915	82	4280	3360	0.74793	105
18	14	725	67	4705	3450	0.73326	103
18	15	655	51	4410	3690	0.83673	98
18	16	670	59	5130	3270	0.63742	105
18	18	715	66	5050	3580	0.70891	95
18	19	705	68	5075	3240	0.63842	91
18	20	610	60	4890	3680	0.75255	94
18	21	665	56	5035	3510	0.69712	89
18	23	680	65	5005	3790	0.75724	91
18	24	730	74	4540	3840	0.84581	81
18	25	725	71	4680	3050	0.65170	87
18	26	830	77	4860	3450	0.70987	83
18	27	870	72	5020	3140	0.62549	90
18	28	890	70	5050	3015	0.59702	84
18	29	845	73	4810	3240	0.67359	79
18	30	905	86	4780	3310	0.69246	82

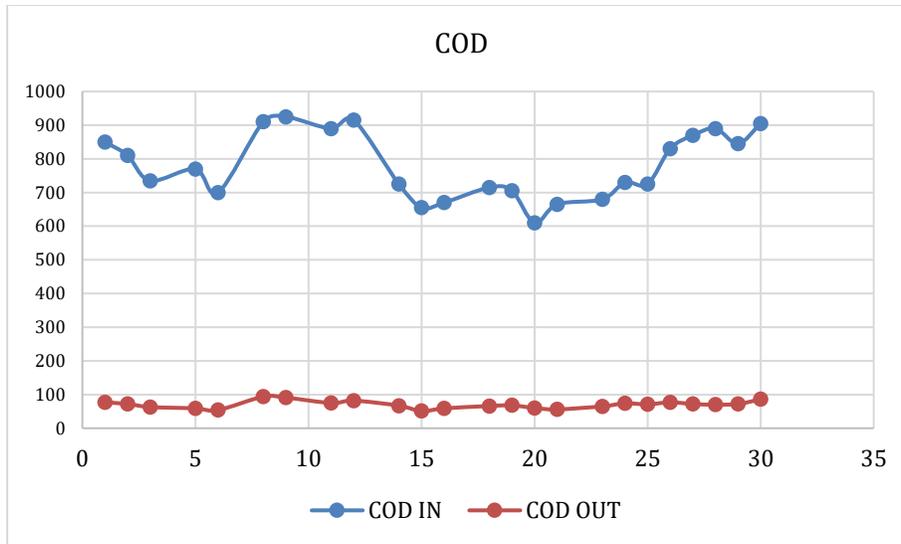


Figure 2: COD changes using real effluent

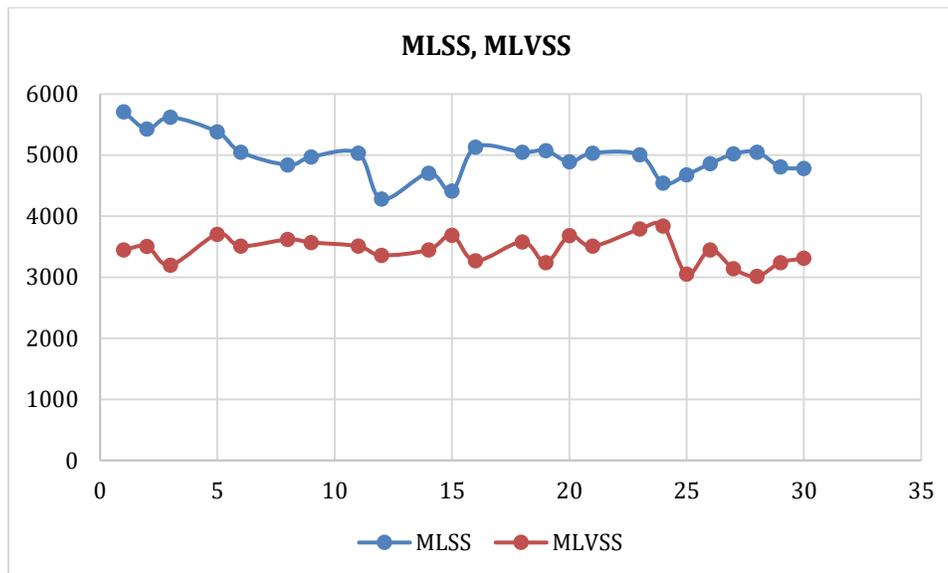


Figure 3: Changes of MLSS and MLVSS using real effluent

Results of SVI tests

The sludge volumetric index (SVI) is a key parameter used to characterize the morphology of activated sludge, along with viscosity, particle size, EPS, and SMP, and microscopic observations. During the first operational phase, SVI decreased roughly. The reduction of SVI indicates the compaction of activated sludge. In addition, due to the infinite SRT in the system, it is expected that the ratio of protein to polysaccharide in the floc will decrease, and as a result, we predict the disintegration of the

aggregates and the increase of free microorganisms.

In general, the disintegration of microbial aggregates and the reduction in the size of flocs can be justified by the fact that because bacteria need to increase the mass transfer surface to consume the substrate, by spreading and becoming smaller, the surface of mass transfer increases and the organic matter. They reach their surface and come in contact with them more easily and can consume organic materials more easily (Figure 4).

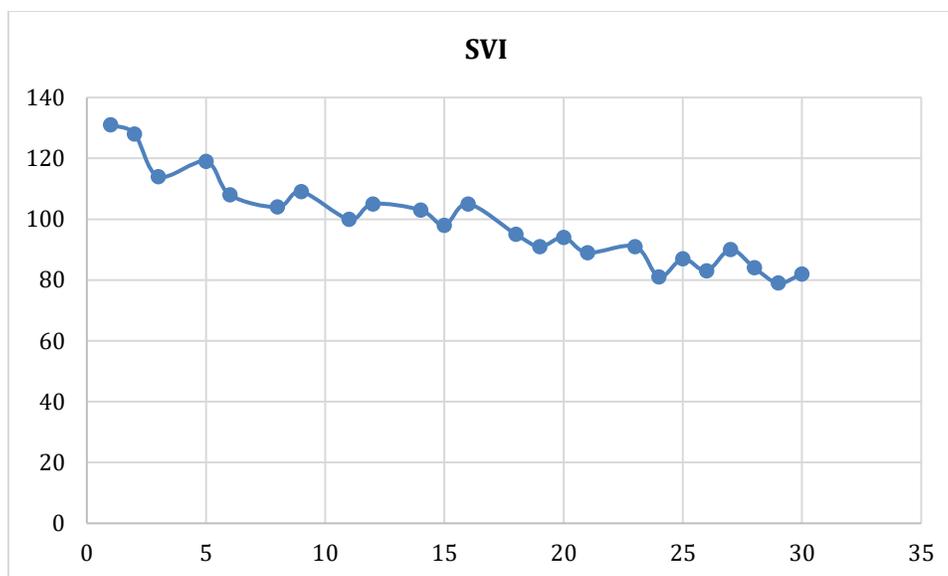


Figure 4: Variations of SVI using real effluent

Conclusion

Due to the limited amount of real effluent to use in this project, the hydraulic retention time (HRT) of 18 hours during one month of operation was considered. The TDS value of this effluent was 1430 at the start of the operation and reached to 1680 after one month. The rise was caused by the addition of nutrients and carbon and phosphorus sources. There is no sludge return to the bioreactor in this project, indicating an indefinite SRT. The hydraulic retention time is the only operational variable in this project. Because the amount of real effluent is limited and is only enough for one hydraulic retention time, synthetic effluent containing a mixture of 93 grams of methanol and 297 grams of pyrolysis gasoline per 1000 liters of water was used to continue the project (after a few months stop for pilot overhaul), which began with inoculation of new activated sludge. Since the new sludge was provided from the same site of the Persian Gulf Fajr Energy Company, the new activated sludge had identical properties, and it was injected into the reactor on January 15, 2022, after concentrating to 5400 ppm. SVI of the inoculated sludge was 130 with the initial concentration of about 2500 ppm. Despite of using 8 used membranes in this section of the project before the overhaul process, acceptable

results were attained. After the overhaul process of the pilot, the 8 used membranes were removed and replaced with 5 new and unused membranes. The pressure difference between the two ends of the membranes was always less than 200 millibar in the early phases of the startup, indicating that the new membranes are not blocked in the startup. The COD value of the synthetic effluent employed in this stage was 580, and the TDS value was 7.37 mg/l. The pilot's COD removal efficiency was excellent, and the output COD was consistently less than 20 mg/L. Consequently, the effluent was standard to be discharged to the environment or used as the makeup water for cooling towers. To investigate the bacterial content of the effluent, the treated effluent from the MBR pilot was entered to the cooling tower pilot as circulating water, and the effect of the biocide number HE0085 developed by Abrizan Industrial Research Company, was investigated (this company has a multi-year cooperation memorandum with a Petrochemical Research and Technology Company). TBC and SRB microbiological tests reveal that the treated water in these circumstances meets the required standard to be used as cooling tower makeup water.

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