Treatment of Real Paper-Recycling Wastewater in a Novel Hybrid Airlift Membrane Bioreactor (HAMBR) for Simultaneous Removal of Organic Matter and Nutrients

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ABSTRACT: *In this study, a novel integrated Hybrid Airlift Membrane Bioreactor (HAMBR) composed of oxic, anoxic and anaerobic zones was developed to simultaneously remove organic matter and nitrogen from real paper-recycling wastewater. The removal efficiencies of Chemical Oxygen Demand (COD), ammonium, nitrite, nitrate and Total Nitrogen (TN) for permeate and supernatant were in the range of 88-99%, 54-83%, 70-90%, 65-95% and 61-90%, respectively. In addition, the membrane fouling was evaluated by Trans-Membrane Pressure (TMP) monitoring during experimental period at a constant flux of 12 L/m^{2}h, and the rate of TMP increase was 1.75 mbar/day. The results showed that the hybrid airlift membrane bioreactor can be applied effectively to simultaneous removal of organic and nutrient from real wastewater and the performance of the membrane bioreactor was satisfactory in terms of resistance against membrane fouling phenomenon, which is an important parameter during HAMBR operation.*

KEYWORDS: *Paper-recycling wastewater; Hybrid Airlift Membrane Bioreactor (HAMBR); Membrane fouling; Organic matter.*

INTRODUCTION

The increase in human population and the demand for industrial establishments have created many problems, such as over-exploitation of resources, which leads to the pollution of the environment. It has been estimated

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1021-9986/20/3/203-211 9/$/5.09

that the pulp and paper industry is responsible for 50% of all wastes dumped into rivers. Water reclamation in pulp and paper industry has been emphasized due to consumption of a large amount of water in this area [1]. More than 250 chemicals have been specified in pulp and paper wastewater and their discharge into the environment can make important environmental problems [2, 3]. This type of wastewater is characterized by high levels of COD, Suspended Solids (SS), color, toxic chlorinated compounds, tannins, resin acids, heavy metals, sulphur compounds, lignin and its derivatives [4-6].

Depending on the process used in pulp and paper mills, different types of treatment technologies, which are efficient and exclusively designed, have been evaluated to minimize the harmful effect of effluent on the environment [7-9]. However, the primary clarification, secondary and tertiary treatment, which can be categorized into physicochemical and biological processes, are the main methods in pulp and paper mills [10]. Although, biological techniques are economical and ecofriendly, treatment of wastes including non-biodegradable recalcitrant compounds mostly limits their broad application. Therefore, other innovative approaches, such as Advanced Oxidation Process (AOP), novel biodegradable polymeric flocculants, electrocoagulation and photo catalysis have been used to tackle this problem [11].

The conventional methods for treatment of paper mill wastewater consist of a balance tank, a first sedimentation tank, an anoxic-aerobic tank and a secondary sedimentation tank. In general, the conventional treatment cannot meet the water quality requirements for paper making process. Consequently, the final effluent includes more than 40% organics with low biodegradability in the total organic matter content. Therefore, the integrated membrane bioreactor has been used to treat the wastewater of paper mills [1, 12]. An MBR combines an activated sludge process with a physical separation by membrane. Compared with Conventional Activated Sludge (CAS) process, the above-mentioned technique has many advantages, including high efficiency, less sludge production, high disinfection ability and also small footprint [13]. One of the main advantages of a membrane bioreactor system is the complete biomass retention, which enables the handle of Sludge Retention Time (SRT) independently of Hydraulic Retention Time (HRT) [14].

There have been some studies on the application of MBR process to treatment of wastewater in pulp and paper industry. It was reported that the HRT of 1.1 ± 0.1 days was an optimal value for COD removal and cake formation was the dominant mechanism of membrane fouling [15]. *Dias et al.* [16] used membrane bioreactor for treatment of kraft pulp mill foul condensates, and the results showed that the treatment at high temperatures was technically feasible and had acceptable potential for industrial applications.

Nevertheless, membrane fouling phenomenon is a major drawback for the widespread application of MBR systems and has been extensively analyzed as a function of operation condition [17, 18]. In this phenomenon, the small and soluble particles penetrate inside the membrane and are adsorbed into the membrane pores along with other organic and inorganic matters, which decreases the permeate flux value to below the theoretical capacity of the membrane filtration and leads to a significant increase in the consumed operational energy [19-21]. Generally, fundamental membrane fouling can be developed over long term operation either by accumulation of particles on the membrane surface or by blockage of the pores. Although significant attempts have been made to gain a better understanding of the dominant fouling mechanisms, control of membrane fouling has been limited because of its complexity. Fouling is influenced by some factors, including membrane pore size, pollution loading rate, particle distribution, membrane material properties and operation conditions. In order to ensure the membranes have an efficient and long operational life, they should be cleaned with physicochemical techniques [21-24].

In aerobic MBRs, approximately complete reduction of organic matters and nitrification can be achieved, while for denitrification process, some modifications are needed, such as the addition of an anoxic tank prior to the aerobic tank, the modification of the reactor configuration including intermittent aeration and baffled membrane bioreactors [25, 26]. It was reported that hybrid membrane bioreactor systems, including sequential or alternating anoxic-oxic zones, have been developed very successfully and activity of the sludge is maintained by various environmental conditions, resulting in effective removal of pollutants [27-29]. In general, evaluation of integrated bioreactors which combine aerobic and anaerobic degradation pathways in a single reactor is important because they are cost-effective and efficient and have smaller footprints as compared to the sequential anaerobic-aerobic systems [30, 31].

The aim of this study is to evaluate the performance of HAMBR in terms of organic compounds and nutrients removal and in addition, analysis of membrane fouling during treatment of real paper-recycling wastewater.

Paper-recycling wastewater

The samples of paper-recycling wastewater used in this study were collected from Kahrizak paper mill located approximately 10 km far from Tehran, Tehran province, Iran. The process of wastewater treatment in this factory was sequencing batch reactor (SBR). The wastewater had COD of 1340-1780 mg/L and SS of 1045-1275 mg/L. The wastewater also contained ammonia (NH_4-N) and TN concentration of 31.2-53.7 mg/L and 137.5-173.5 mg/L, respectively. The pH value of the wastewater was 7.3-7.7.

HAMBR reactor set-up and experimental process

The configuration and operating conditions of HAMBR are all demonstrated in Fig. 1 and Table 1. The membrane bioreactor was operated for 40 days without discharging any excess sludge, except for small amounts (300 mL) for sampling and analysis. The flux was kept constant through frequently adjusting the rotation rate of the suction pump, and water level sensor was used to maintain the constant water level in the HAMBR.

There was a recirculation pump which worked with recirculation of around 400% of the inflow. A Kubota A_4, a flat sheet microfiltration membrane made of chlorinated polyethylene (area 0.106 m^2, pore size 0.4 μm), was used in the bioreactor. Air was supplied through a diffuser under the flat sheet membrane module in order to provide oxygen for activated sludge, the driving forces for the circulation of the suspension inside the HAMBR and the membrane scouring. The pressure gauge was installed between the membrane and the effluent pump in order to monitor the variation of the TMP [32, 33]. The bottom of HAMBR was packed by Granular Activated Carbon (GAC) media in order to develop biofilm and increase removal efficiency of pollutants [34]. The performance of the bioreactor for removal of pollutants was analyzed through several conventional indexes of water quality, including COD, NH^+_4, NO^-_2, NO^-_3 and TN.

Table 1: Operating conditions of the HAMBR.

Parameter	Value
HRT (h)	36
pH	7-7.5
SRT (day)	up to 40
OLR (kg COD/m^3day)	0.89-1.2
Temperature (ºC)	20-25

Analytical methods

The concentrations of COD, NH^+_4, NO^-_2, NO^-_3 and TN for both supernatant and permeate were analyzed using specific test kits on a Hach DR 5000 Spectrophotometer (Loveland, CO). The concentration of SS was determined in accordance with the Standard Methods for the Examination of Water and Wastewater [35]. The pH value was measured using a pH meter (691 pH Meter, Metrohm). The TMP was monitored by a pressure gauge set between the membrane module and the suction pump.

RESULTS AND DISCUSSION

Organic carbon removal

Fig. 2 shows the variation of COD concentrations and removal efficiencies during the operating time. The COD concentration in permeate (COD_p) and supernatant (COD_s) remained lower than 65 mg/L and 175 mg/L, respectively, during the experiment, and an average reduction of 98% and 95% was observed, respectively. While COD removal in a conventional MBR was reported to be 80% [36] and at a hybrid moving bed biofilm reactor-membrane bioreactor (hybrid MBBR-MBR) containing carriers, it was 85.82 ± 2.12% [37]. Additionally, for a jet loop MBR which was fed by olive mill wastewater, approximately 93% removal efficiencies in terms of COD was achieved [38].

The COD concentration of supernatant (COD_s) was higher than COD_p, which shows the beneficial effect of dynamic membrane for increasing the COD removal and the fact that this COD is a consequence of biological

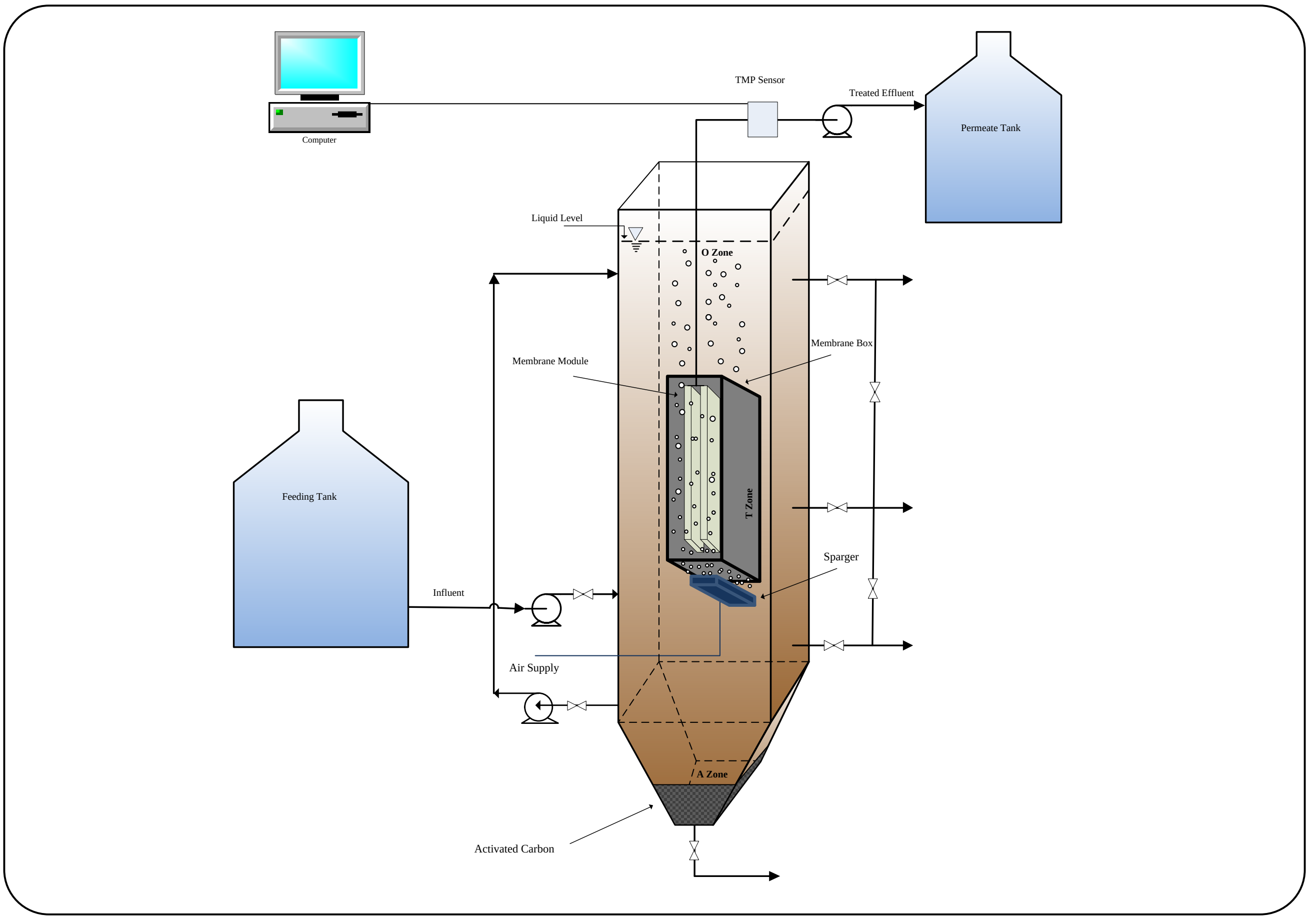


Fig. 1: Schematic diagram of the HAMBR.

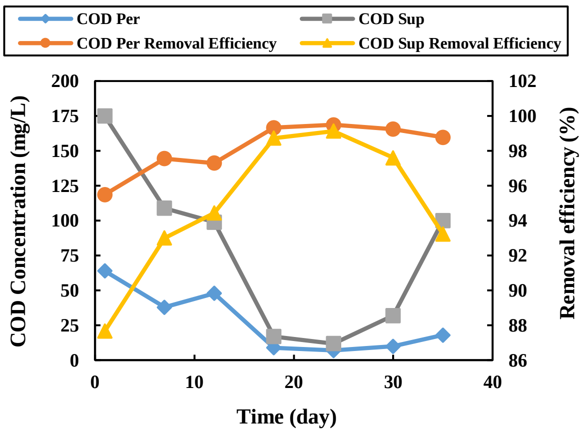


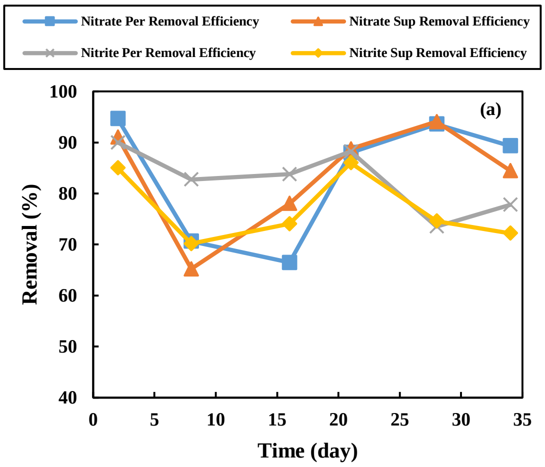
Fig. 2: COD concentration value and removal efficiency in permeate and supernatant versus time in the HAMBR.

activity [32]. These findings were in line with other studies [39, 40]. Nevertheless, the COD_s removal value had more fluctuation compared with COD_p. This fluctuation was due to the change of concentration of pollutants in the real wastewater. The significant reduction in COD shows that the continuous supply of organic matters in the feed is utilized by the microbial population. This is supported by the growth in MLSS during continuous operation [41].

Nitrogen removal

Fig. 3 demonstrates the ammonium, nitrite, nitrate and TN removal efficiencies during the experimental period of HAMBR. Totally, converting nitrogenous substances to nitrogen gas needs an oxic and anoxic environment to create an appropriate nitrification-denitrification condition. Nitrite is the product of ammonia oxidation and a connecting point between nitrification and denitrification and can convert not only to nitrogen gas but also to nitrate via nitrification. Such coordination varies depending on the culture conditions and bacterial population [42].

Fig. 3 (a) indicates the concentration removal of nitrite and nitrate. It was observed that nitrite removal efficiencies were in the range of 70-90%, indicating that most nitrite has been converted and nitrification has taken



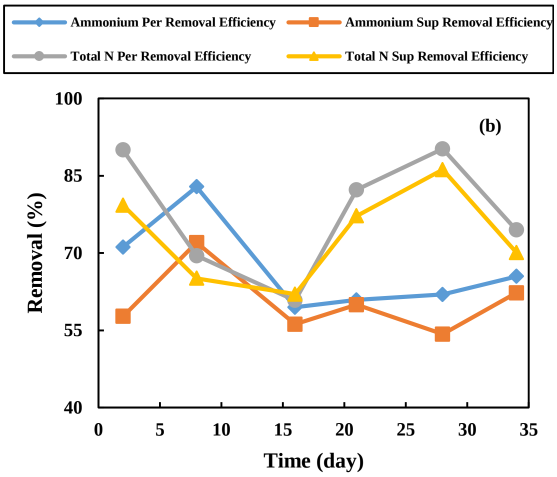


Fig. 3: Removal efficiency of the HAMBR: (a) Nitrite and Nitrate and (b) Ammonium and TN.

place. In addition, nitrate removal was more than 65%, which is an acceptable value.

Fig. 3 (b) demonstrates ammonia and TN removal efficiencies during HAMBR operation. Ammonia concentration decreases very obviously and the maximal NH_4-N removal percentage was 83%, indicating that the nitrifying bacteria gradually accumulate in the bioreactor. In addition, in this study, the range of TN removal was 61-90%, while in another study, the airlift MBR was used for simultaneous nitrification-denitrification and the average total nitrogen removals were in the range of 39.4-63.1% [43]. Furthermore, *Kimura et al.* [44] reported that TN removal rate by baffled membrane bioreactor was 77%.

On the other hand, generally, removal efficiencies of nitrogenous substances in permeate were higher than supernatant, which indicates the effectiveness of membrane on removal of nutrients. These results all indicate that the HAMBR was stable and feasible and the treatment efficiency was satisfactory.

Membrane performance

Characterization of fouling during the operation of HAMBR was monitored by TMP. Fig. 4 demonstrates the variation of TMP and flux values during more than 40 days of operation of the HAMBR. The rate of change in TMP value is a significant factor to evaluate the system performance in an MBR system [45], because, at a constant rate of permeate flow, TMP is directly connected to the rate of membrane fouling. The value of permeate flux was maintained approximately 12 L/m^{2}h, and the rate of TMP increasing was 1.75 mbar/day.

As it is obvious in Fig. 4, during the initial 5 days of operation, the increase in TMP of the membranes was relatively great, approximately 6 mbar/day, partly as a consequence of the development of biological activities and bio-accumulation on the membranes. Subsequently, there was a modest increase with some fluctuations in TMP value in 40 days. During operational period, there was not the tendency of a “TMP jump” as reported in many literatures [46, 47], which indicates acceptable performance of membrane and resistance against fouling. The value of TMP reached 163 mbar at the end of the operation compared to 93 mbar at the beginning.

Industrial application and economic evaluation

It was expected that, by 2019, more than 5 million m^3/day of wastewater would be treated by MBR plants in the world. The Henriksdal wastewater treatment plant in Stockholm will be upgraded with an MBR, which will be able to treat 864,000 m^3/day of wastewater, making it the largest MBR plant in the world. In 2004, when the Nordkanal MBR plant was commissioned, it was the largest MBR plant with a design capacity of 45,000 m^3/day. This increase in the design capacity between the Nordkanal and the Henriksdal plants shows the significant growth of MBR technology [48, 49].

Energy demand and membrane fouling are important features regarding economic aspects of MBR. For this reason, these factors are focal points in the full-scale MBR

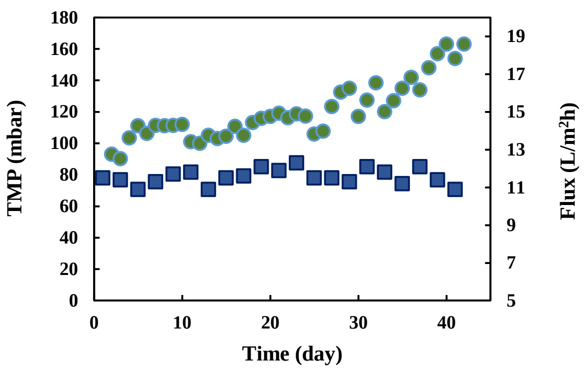


Fig. 4: TMP and flux profile during operating period.

design and operation [49]. According to the recently reported data from full-scale MBRs, the average annual specific energy consumption varies between: 0.8-2.4 kWh/m^3 in France [50], 0.8-3.0 kWh/m^3 in Japan [51], 0.4-0.6 kWh/m^3 in China [52] and 0.4-2.1 kWh/m^3 in Spain [53]. For this reason, recent developments in membrane bioreactor energy reduction focused on the module configuration, aeration strategies, control systems, low-energy membrane cleaning methods or novel fouling mitigation methods [18, 54]. On the other hand, since membrane prices have significantly decreased during the last 15 years, MBR technology has become a more attractive solution for medium sized plants with a population equivalent of 10,000-100,000. Furthermore, considerable progress has been accomplished in the design and operation optimization of MBR systems, which has helped to reduce the capital and operating expenses of MBR plants. The global market for MBRs was $425.7 million in 2014 and is projected to approach $777.7 million by 2019, registering a Compound Annual Growth Rate (CAGR) of 12.8% in the period 2014-2019 [49].

CONCLUSIONS

Through an HAMBR experiment lasting 40 days under the provided operational conditions, removal of pollutants was evaluated. It was concluded that multi-zone conditions form gradually in the single membrane bioreactor, creating a beneficial environment for simultaneous nitrification and denitrification for the removal of the carbonaceous and nitrogenous materials, and permeate quality reached below the standard of Iran. The removal efficiencies were achieved through an HRT of 36 h, and the best COD removal efficiency was 99%. In addition, the performance of the system was acceptable in terms of resistance against fouling phenomenon, and the TMP development was only 1.75 mbar/day, which is an important parameter during HAMBR operation.

Acknowledgements

This study was financially supported by grant No: 950704 of the Biotechnology Development Council of the Islamic Republic of Iran.

Conflict of interest

The authors would like to declare that there is no conflict of interest associated with this research and its publication.

Received : Nov. 18, 2018 ; Accepted :Mar. 11, 2019

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