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Membrane Filtration as Post-Treatment of Rotating Biological Contactor for Wastewater Treatment

Sharjeel Waqas [1,2], Muhammad Roil Bilad [3,4,*], Nurul Huda [5,6,*], Noorfidza Yub Harun [1], Nik Abdul Hadi Md Nordin [1], Norazanita Shamsuddin [4], Yusuf Wibisono [7], Asim Laeeq Khan [8] and Jumardi Roslan [5]

1 Chemical Engineering Department, Universiti Teknologi PETRONAS, Bandar Seri Iskandar 32610, Perak, Malaysia; sharjeel_17000606@utp.edu.my (S.W.); noorfidza.yub@utp.edu.my (N.Y.H.); nahadi.sapiaa@utp.edu.my (N.A.H.M.N.)
2 School of Chemical Engineering, The University of Faisalabad, Faisalabad 37610, Pakistan
3 Faculty of Applied Science and Technology, Universitas Pendidikan Mandalika, Jl. Pemuda No. 59A, Mataram 83126, Indonesia
4 Faculty of Integrated Technologies, Universiti Brunei Darussalam, Jalan Tungku Link, Gadong BE1410, Brunei; norazanita.shamsudin@ubd.edu.bn
5 Faculty of Food Science and Nutrition, Universiti Malaysia Sabah, Jalan UMS, Kota Kinabalu 88400, Sabah, Malaysia; jumardi@ums.edu.my
6 Department of Food Science and Technology, Faculty of Agriculture, Universitas Sebelas Maret, Surakarta 57126, Indonesia
7 Department of Bioprocess Engineering, Faculty of Agricultural Technology, Brawijaya University, Malang 65141, Indonesia; y_wibisono@ub.ac.id
8 Department of Chemical Engineering, COMSATS Institute of Information Technology, Lahore 54000, Pakistan; alaeeqkhan@cuilahore.edu.pk
* Correspondence: roil.bilad@ubd.edu.bn (M.R.B.); drnurulhuda@ums.edu.my (N.H.)

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Abstract: A rotating biological contactor (RBC) offers a low energy footprint but suffers from performance instability, making it less popular for domestic wastewater treatment. This paper presents a study on an RBC integrated with membrane technology in which membrane filtration was used as a post-treatment step (RBC–ME) to achieve enhanced biological performance. The RBC and RBC–ME systems were operated under different hydraulic retention times (HRTs) of 12, 18, 24, and 48 h, and the effects of HRT on biological performance and effluent filterability were assessed. The results show that RBC–ME demonstrates superior biological performance than the standalone RBC. The RBC–ME bioreactor achieved 87.9 ± 3.2% of chemical oxygen demand (COD), 98.9 ± 1.1% ammonium, 45.2 ± 0.7% total nitrogen (TN), and 97.9 ± 0.1% turbidity removals. A comparison of the HRTs showed that COD and TN removal efficiency was the highest at 48 h, with 92.4 ± 2.4% and 48.6 ± 1.3% removal efficiencies, respectively. The longer HRTs also lead to better RBC effluent filterability. The steady-state permeability increased respectively by 2.4%, 9.5%, and 19.1% at HRTs of 18, 24, and 48 h, compared to 12 h. Our analysis of membrane fouling shows that fouling resistance decreased at higher HRTs. Overall, RBC–ME offered a promising alternative for traditional suspended growth processes with higher microbial activity and enhanced biological performance, which is in line with the requirements of sustainable development and environment-friendly treatment.

Keywords: attached growth process; biological treatment; biofilm; membrane fouling; rotating biological contactors; wastewater treatment

1. Introduction

The treatment of wastewater using biological processes is an economical, energy-efficient, and environmentally sound approach [1]. Typically, microbial aggregates are employed to biodegrade the organic compounds and nutrients in the wastewater [2]. Suspended flocs (employed in the conventional activated sludge (CAS) process) and attached biofilm (employed in trickling filters and rotating biological contactor (RBC)) are the two types of aggregates. Both the suspended flocs and the attached biofilm processes are established in full-scale wastewater treatment processes [3]. RBC—also referred to as biofilm reactors—offers a substitute to the CAS process [4].

There are two main types of RBC configurations: integral and modular [5]. The integral system, a compact decentralized unit, consists of a single unit combining the primary treatment, the RBC bioreactor, and the final clarifier. The integral units are usually contained within a package plant and have treatment capacities of ≤250 population equivalents (PE) [6]. On the other hand, the modular systems have separate independent operations for the primary treatment, the RBC bio-zone, the clarifier, and the solids treatment unit. Modular systems are centralized networks that allow for more flexible process configurations and have treatment capacities of >1000 PEs [7].

RBCs are equipped with solid media as a platform for the development of microorganisms as a biofilm [8]. Unlike the CAS process, which uses air bubbling to supply oxygen in the suspended flocs, the RBC employs mechanical rotations of the disks for contacting the biofilm adhered to them with air to provide the oxygen [9,10]. The commercial significance of membrane technology in municipal and industrial wastewater treatments is increasingly pervasive [11,12]. RBC systems have the inherent drawback of a high footprint due to space requirements for air contact and modular construction. Therefore, the application of such a system must be adjusted to meet these constraints. It is an attractive choice to facilitate the reuse of municipal and industrial wastewaters. The integration of membrane processes has been proven to be superior compared to conventional biological processes [13].

RBCs are seen as a low-cost biological process and an alternative to the more established CAS in treating wastewater [14,15]. The biofilm in RBC is, to a certain degree, resistant to toxic compounds, shock in organic loadings, and moderate changes in hydraulic loading. Biofilm also maintains a high proportion of microorganisms to ensure a high pollutant removal capacity [16,17].

Despite offering the aforementioned benefits, RBCs still suffer from operational instability and variable effluent quality, which are attributed to the biofilm dynamics and the detachment of the biofilm. To overcome these issues, various RBC configurations, including RBC/biofilm, RBC/suspended growth combination, RBC/wetland combination, and RBC membrane technology systems have been proposed [18,19]. RBCs can be installed modularly in parallel, allowing for more stable loadings. If the effluent quality is of primary interest, it can be operated in a series. Hybrid systems can also be considered where an RBC is combined with another unit to enhance the combined process stability to cater to variable loadings, boost the load capacity, or enhance the attainable effluent standard [5]. RBCs can be combined with a polishing step (tertiary treatment) or the capacity can be upgraded [20]. For instance, the RBC/wetland configuration provides a storm flow buffer and improves the discharge effluent quality [7]. In previous studies, the application of membrane technology in combination with RBCs as a post-treatment helped to enhance effluent quality and removal efficiency [21,22].

Studies with RBC systems have revealed that longer contact times improve the diffusion of the substrate into the biofilm and its consequent removal of the influent. This trend is also verified with toxic and heavy metals substrates [23]. The hydraulic retention time (HRT) is directly related to the organic and hydraulic loading of the influent wastewater. Longer HRTs facilitate the proper degradation of the substrate substances and improve removal efficiencies. The trend remains the same for toxic and heavy metal substrates. A short HRT would result in a poor treatment process due to the improper degradation of the substrate, while longer HRTs would affect the process economics. The selection of the optimum HRT is very important for obtaining the desired effluent quality at a minimal cost [24,25]. Najafpour et al. [8] studied the effect of HRTs on the performance of RBCs for the treatment of palm oil mill effluents and found that both the COD (45 to 88% removal efficiency) and total nitrogen (TN) removal efficiency increased with an increase in the HRT (10–55 h). The impact of the HRT is more for the 10–26 h than the 26–55 h interval change.

Ghalehkhondabi et al. [26] studied that chemical oxygen demand (COD) and ammonium removal rates increased with HRT, and the maximum removal rate depends on the selected HRT and the interactions between the disks' rotational speeds.

Saha et al. [13] showed that the combination of RBC with an external membrane for the treatment of oily wastewater resulted in higher treatment efficiency than a stand-alone RBC at all feed ratios and HRTs. A total petroleum hydrocarbon removal efficiency of 99% was obtained at a 24 h HRT in an RBC system combined with an external membrane bioreactor. Similarly, the micro-pollutant degradation and general treatment efficiency could be improved by altering the HRTs. According to Win et al. [27] and Zhang et al., [28], HRT changes affect the bacterial communities and thus, the system performance. Overall, it has been demonstrated that integrating an RBC with membrane technology increases the overall system performance. The attached biofilm in the RBC, with a higher biomass concentration and microbial diversity, favors the integration of membrane separation because of less contact between the biomass with the membrane, which may otherwise cause membrane fouling.

Instigated by the success of CAS incorporated into the membrane filtration, an RBC bioreactor combined with membrane filtration (RBC–ME) is proposed in this study. RBC–ME combines the conventional RBC with a membrane filtration unit placed externally on the bioreactor. This study evaluates the integration of an RBC system with membrane filtration by replacing the settling tank with a membrane filtration unit. In previous studies, the membrane filtration was placed after the settling tank in a separate vessel, which required extra space. The main functionality of an RBC–ME is the membrane placement into the settling tank immediately after the RBC bioreactor. The integration of the membrane with an RBC is expected to maintain high effluent quality and removal efficiency. The HRT is an important operation parameter and an optimized valve has a strong interaction with the effluent quality and plant economy. Limited literature is available for an RBC integrated with membrane technology; hence, the current study aims to study the effect of HRT on the COD and ammonium removal along with membrane permeability.

This study focused on the process intensification of the RBC–ME, particularly on the biological performance and the membrane fouling control. The operational parameter of the HRT was selected for this study, as it is one of the most influential parameters in the bioreactor. Alterations in HRTs could also result in variations of organic loading rates. Firstly, the biological performances, in terms of organic and nutrient compound removal of both standalone RBC and RBC–ME, were evaluated as a function of the hydraulic retention time. The membrane filtration and fouling analysis are presented at various HRTs.

2. Materials and Methods

2.1. Wastewater Preparation and Characterization

Synthetic wastewater was prepared by blending refined food leftovers (1 g/L), as suggested in previous work [29]. After mixing food leftovers with water, the mixture was left for 2 h to settle the suspended particles. The stock solution (supernatant) was then filtered through Whatman filter paper, 11 µm medium flow filter paper (Grade 1 Qualitative Filter Paper Standard Grade, GE Whatman, Kent, UK). The stock solution was then diluted to obtain the influent wastewater concentration, as summarized in Table 1. The prepared wastewater was analyzed in terms of COD, TN, ammonium, and nitrate.

Table 1. Influent characteristics for the RBC and RBC–ME bioreactors.

	Influent
COD (mg/L)	298 ± 45.6
TN (mg/L)	2.4 ± 0.2
Ammonium (mg/L)	0.92 ± 0.07
Nitrate (mg/L)	0.52 ± 0.08
Turbidity (NTU)	15.2 ± 0.6
pH	6.35 ± 0.18

RBC: rotating biological contactor, RBC–ME: rotating biological contactor–membrane external, COD: chemical oxygen demand, TN: total nitrogen.

The COD, the TN, the ammonium, and the nitrate were measured using the specific Hach digestion solution (HACH, Loveland, CO, USA) for each compound. The solution was diluted accordingly to fall into the range of the digestion vials. The values were determined through a Hach DR3900 Spectrophotometer (HACH, Loveland, CO, USA). A Hach 2100Q portable turbidimeter (HACH, Loveland, CO, USA) and a Hach HQ411D benchtop PH/MV meter (HACH, Loveland, CO, USA) were used to determine the turbidity and pH, respectively [30].

2.2. Membrane Preparation and Characterization

A polysulfone (PSF) membrane was prepared through the phase inversion technique detailed in Table 2. The PSF membrane was fabricated from a dope solution containing PSF as the polymer, (BASF-Ultrason, Mw 22 kDa), polyethylene glycol (PEG) as the additive (Sigma-Aldrich, MO, USA, Mw 10 kDa), and *N*,*N*-Dimethylacetamide (DMAc) as the solvent (Sigma-Aldrich, MO, USA) at concentrations of 12, 1, and 87 wt%, respectively. The dope solution was cast according to the phase inversion method, as described in our previous study [19]. The cast film was immersed immediately in demineralized water (acting as a non-solvent) to form a membrane sheet. A solid, thin, and porous membrane was then stored in water until further usage.

Table 2. Summary of materials for membrane fabrication.

IUPAC Name	Abbreviation	Avg. Molecular Mass	Purity *
Polysulfone	PSF	22,000 Da	100 wt%
Polyethylene glycol	PEG	9000–12,500 Da	100 wt%
N,*N*-Dimethylacetamide	DMAc	-	99.7 vol%
Water	H_2O	-	~100 vol%

* vol% = by volume percentage, wt% = by weight percentage.

The properties of the fabricated PSF membrane are listed in Table 3. The membrane thickness was measured using an electronic digital micrometer screw gauge (Mitutoyo 293-340-30 Digital Micrometer, Mitutoyo America Corporation, Aurora, CO, USA). The pore size was measured using a capillary flow porometer (Porolux, Nazareth, Belgium). The morphology of the membrane was analyzed, and the microstructure was acquired using scanning electron microscopy (SEM) (Zeiss, Leo 1430 VP, Carl Zeiss, Oberkochen, Germany). The static membrane surface water contact angle was determined using the sessile drop method (Mobile Surface Analyzer, KRUSS, Hamburg, Germany). All the characterization techniques were done in triplicate.

Table 3. Summary of membrane properties used in RBC–ME configuration.

Properties (Unit)	Values
Materials	Polysulfone
Thickness (mm)	0.28 ± 0.22
Mean flow pore size (μm)	0.03 μm
Surface contact angle (°)	61.8 ± 1.0
Cross-section morphology	Asymmetric
Clean water permeability (L/(m^2 h bar)	817 ± 35

2.2.1. Determination of Filtration Performance

The membrane filtration was done at a fixed transmembrane pressure (ΔP) of 0.1 bar with a system detailed elsewhere [31]. The low pressure for filtration not only reduces the energy cost but it is also less susceptible to membrane fouling and maintains sustainable flux, as reported elsewhere [32]. The membrane permeability (L, L/m^2 h bar) was calculated using Equation (1).

$$L = \frac{\Delta V}{A\ \Delta t\ \Delta P} \quad (1)$$

where V is the volume of permeance (L), A is the membrane area (m^2) and t is the filtration time (h). During the filtration test, the permeate pump was stopped temporarily and the filtration was run in a full recycle without altering the hydraulic operation parameter.

2.2.2. Membrane Fouling Analysis

The membrane fouling analysis was evaluated using the Darcy law detailed in Equations (2) and (3).

$$J = \frac{\Delta P}{\mu R_T} \quad (2)$$

$$R_T = R_M + R_P + R_C \quad (3)$$

where J is the permeate flux, μ is the dynamic viscosity of the permeate, R_T is the total resistance, R_M is the intrinsic membrane resistance, R_P is the pore blocking resistance, and R_C is the cake layer resistance. The R_M was calculated by the filtration of deionized water and R_{M+P} was calculated from the filtration of deionized water using the used membrane after removing the cake layer from the membrane surface.

2.3. Bioreactor Set-Up and Operation

The lab-scale RBC–ME bioreactor was fabricated in-house (Figure 1). It comprised a feed tank, a bioreactor tank, and a settling tank. The bioreactor unit was a cuboid of $25 \times 25 \times 30$ cm, fabricated using acrylic sheets. It had a total working volume of 6.5 L, with a 40% disk submergence. The unit consisted of 5 rotating disks (D = 18 cm) separated by a 3 cm gap between adjacent disks and fixed on a stainless-steel shaft. The rotating disk had a total surface area of 2034 cm^2. Polyurethane sheets were cut according to the size of the disks and then glued on the disks to be used as a platform for biofilm growth. The activated sludge used to inoculate the lab-scale RBC–ME was obtained from a nearby full-scale activated sludge domestic wastewater treatment plant.

The membrane sheet was cut and fixed onto both sides of the panel onto a plate and frame filtration panel. The membrane sheet was attached to a panel consisting of a semi-circle shape and resulted in an active membrane surface area of 226 cm^2. The membrane sheets were glued to the panel with AB epoxy glue (AB quick epoxy, HYRO, Kuala Lumpur, Malaysia). The filtration panel was ensured to be free from leakage. A spacer fabric that was placed between the two membrane sheets acted as permeate channel. The membrane permeate was evacuated through a permeate pipe that connected the permeate channel to the permeate pump.

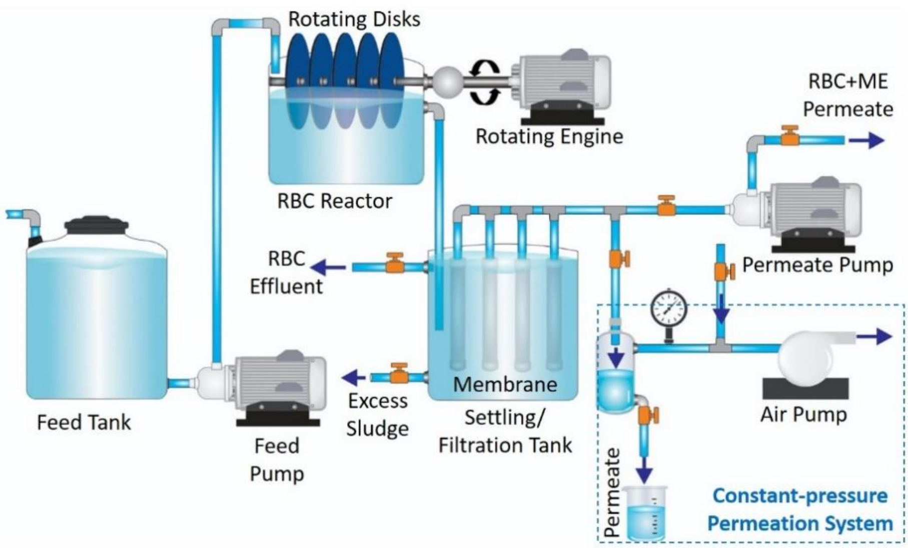


Figure 1. Schematic diagram of the laboratory-scale RBC and RBC–ME unit.

There were 2 different bioreactor configurations, one with membrane filtration and the other without membrane filtration. In the first configuration, the stand-alone RBC bioreactor followed by a settling tank was used to degrade the organics and nutrients. The treated water was flown by gravity to the settling tank and from the settling tank to a sink. The HRT was adjusted from the feed pump. In the second configuration, membrane filtration was incorporated and placed in the setting tank to allow filtration externally. This configuration is henceforth referred to as RBC–membrane external (RBC–ME). The membrane in this configuration acts as a post-treatment. The RBC–ME configuration eliminated the need for a settling tank, and thus, the addition of membrane filtration does not alter the overall plant size. To adjust the HRT in the RBC–ME, a peristaltic pump was installed to drive the permeation accordingly to meet the required HRTs.

The RBCs were run for 42 days, divided into 2 periods. During the first 15 day period, the bioreactor was operated under constant loading conditions of 17 g COD/m^2.d to grow and acclimatize the biofilm atop of the polyurethane foam surface. During this period, the biofilm was observed carefully, and the biological performance was monitored regularly. Carbonaceous bacteria responsible for COD biodegradation were expected to dominate the biofilm, as compared to nitrifying bacteria, which undergo TN removal. After the acclimatization phase, the biofilm was completely developed and was effective in degrading organics and nutrients. Any detached flocs from the rotating disks were regularly discharged.

The 2 system configurations were assessed: (1) the stand-alone RBC bioreactor and (2) the membrane placed externally on the settling tank. In the second phase of the RBC operation, the bioreactor was operated by incorporating the membrane to study the biological and filtration performance. For the RBC–ME configuration, there were no forms of membrane fouling control techniques (coarse bubble aeration, tweaking of hydrodynamics conditions) applied to annihilate membrane fouling. The feed for the RBC–ME had undergone biological treatment in the RBC bioreactor and was expected to pose low fouling potential. The effects of 2 different bioreactors (RBC and RBC–ME) on the biological and hydraulic filtration performance were then assessed.

2.3.1. Hydraulic Retention Time

The performance of both the bioreactors (RBC and RBC–ME) was assessed by varying HRTs from 12 to 48 h at a constant influent wastewater concentration. The performance of the bioreactors, in terms of biological treatment and membrane permeability, was assessed. As the HRT is directly associated with the hydraulic loading rate (HLR), this paper only discusses the effect of HRT. The bioreactor was acclimatized to a 9 h HRT, equivalent to an organic loading rate of 17 g COD/m^2.d and a 67.9 L/m^2.d of HLR. After acclimatization, the bioreactor was operated at a 12 h HRT, equivalent to a 51 L/m^2.d of HLR. The HRT was increased from 12 to 18, 24, and 48 h to determine the effects of COD, TN, turbidity, and membrane permeability.

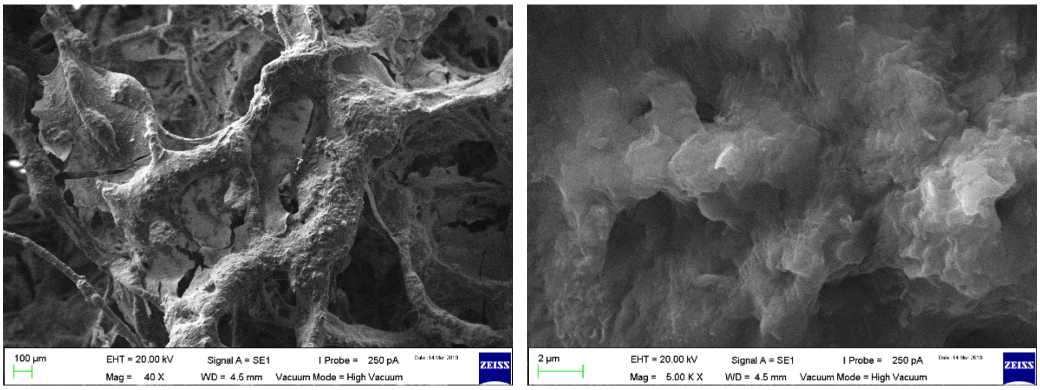
2.3.2. Scanning Electron Microscope

After the acclimatization stage, a 1 cm^2 piece of biofilm was analyzed using SEM analysis. The biofilm sample was carefully cut from the rotating disk. The foam was then treated with formaldehyde for biofilm impregnation according to the method detailed earlier [33] to maintain the biofilm structure. The biofilm sample was then dehydrated by consecutive immersions in 20, 40, 60, 80, and 100% ethanol solution, each step for 5 min, to avoid shrinkage, followed by a drying process. The dried non-conductive sample was sprayed with conductive gold nanoparticles using an ion sputter instrument to create a conductive layer on the sample that reduces thermal damage, inhibits charging, and improves the secondary electron signal required for topographic examination in the SEM. The conductive biofilm sample was loaded onto the SEM sample stage under vacuum conditions and an electron gun shot out a beam of high-energy electrons.

3. Results

3.1. Biofilm Analysis

Figure 2 shows the biofilm developed at the surface of the disk visualized using SEM. The SEM images were obtained at 40× and 5000× magnification levels. Figure 2a shows a birds-eye view of the biofilm established on the carrier media at 40× magnification. The SEM images show the well-established biofilm of microorganisms on the media surface.



(**a**) (**b**)

Figure 2. SEM results of biofilm developed at the surface of the rotating disk under (**a**) 40× and (**b**) 5000× magnifications.

It can be identified as a mature biofilm that occupied the sponge media surface, ascribed to its excellent biological performance in removing organics from the wastewater (detailed in Section 3.3). A mature biofilm with a characteristic mushroom formed of polysaccharides can be seen in Figure 2b. At this stage, cells start to detach and revert to planktonic cells that stick to the new surface to develop another biofilm layer.

3.2. Membrane Characterization

The properties of the applied membrane in the external filtration system are summarized in Table 3. The thickness and mean flow pore size were 0.28 ± 0.22 mm and 0.03 µm, respectively (Figure 3). The sizes of the microorganism species were much larger than the mean flow pore size of the membrane combined with the asymmetric nature of the morphology, thus ensuring complete biomass retention at the membrane surface. For an asymmetric phase-inverted membrane, the membrane pore size is dictated by the size of the pore mouth [34], which, in this context, disallows the penetration of any free biomass into the membrane structure. This advantage ensures no biomass is carried forward to the effluent, nor any suspended matter typically vulnerable in a standard settling system. However, colloidal particles and dissolved nutrients can pass through the membrane pores unless an additional layer of biofilm grows on the membrane surface, which aids in biodegradation, as often occurs in a membrane bioreactor [33]. It is worth noting that the biological performance was less affected by the membrane properties. The membrane samples were asymmetric, as revealed from their cross-section SEM image (Figure 3). The membrane surface water contact angle determines the hydrophilic/hydrophobic nature of the membrane. The membrane surface water contact angle of 61.8 ± 1.0° revealed a hydrophilic membrane. The membrane exhibited a clean water permeability of 817 ± 35 L/(m^2 h bar).

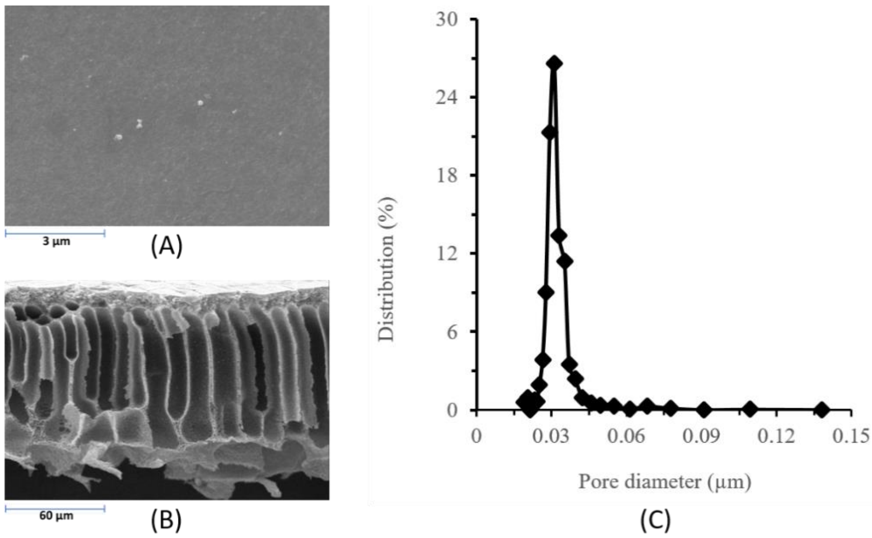


Figure 3. The surface (**A**) and cross-section SEM images (**B**), as well as its pore size distribution (**C**) of the applied membrane in the RBC–ME.

3.3. Biological Performance

Table 4 summarizes the biological performance of RBC and RBC–ME bioreactors for synthetic wastewater. Superior biological performance of the RBC incorporating the membrane showed the significance of membrane integration with the RBC. The results in Table 4 suggest that after the acclimatization period, the bioreactor stabilizes, which is depicted in the steady removal efficiencies. The bioreactors in both the RBC and RBC–ME depicted excellent removal efficiencies for COD, the ammonium, and turbidity. They also showed good performance in maintaining the pH around the neutral value.

Table 4. Effluent characteristics for the RBC and RBC–ME bioreactors employing the PSF membrane.

	RBC Effluent	RBC % Removal Efficiency	RBC–ME Effluent	RBC–ME % Removal Efficiency
COD (mg/L)	78.2 ± 7.5	72.4 ± 2.5	35 ± 8.9	87.9 ± 3.2
TN (mg/L)	1.54 ± 0.05	38.3 ± 1.9	1.41 ± 0.05	45.2 ± 0.7
Ammonium (mg/L)	0.03 ± 0.01	95.6 ± 0.8	0.01 ± 0.01	98.9 ± 1.1
Nitrate (mg/L)	1.9 ± 0.3	-	1.8 ± 0.2	-
Turbidity (NTU)	3.3 ± 0.3	78.9 ± 0.3	0.32 ± 0.03	97.9 ± 0.1
pH	6.82 ± 0.03	-	6.95 ± 0.11	-

RBC: rotating biological contactor, RBC–ME: rotating biological contactor–membrane external, COD: chemical oxygen demand, TN: total nitrogen.

The excellent biological performance in both systems can be explained as follows. Carbonaceous bacteria are responsible for the biodegradation of organic compounds, aerobically using dissolved oxygen as a terminal electron acceptor, while nitrogenous bacteria decompose the nitrogen compounds. Nitrification, an aerobic process, is a two-step process involving the oxidation of ammonium to nitrite through ammonia-oxidizing bacteria (AOB) and then the conversion of nitrite to nitrate through nitrite-oxidizing bacteria (NOB) [35]. The RBC develops abundant AOB and NOB throughout the biofilm along with carbonaceous bacteria. Nitrification occurred in the RBC without encountering any biofilm problems, while exhibiting a low biomass yield and very high sludge ages. The RBC exhibited excellent ammonium removal efficiency throughout the experimentation period. Treatment of wastewater containing a high organics concentration is typically dominated by heterotrophic bacteria that significantly diminishes nitrifier growth. Therefore, nitrogen removal occurs after organics removal during the last stage of the RBC bioreactor [36].

The microbial-rich RBC bioreactor contains a large population of microorganisms. The most abundant phyla found in the biofilm are *Proteobacteria* and *Bacteroidetes*, accounting for two-thirds of the microbial community. The oxygen-rich outer layer of the biofilm contains more *Proteobacteria*, *Bacteroidetes,* and *Nitrospira* than the inner layer [37]. The AOB and NOB are found both in the inner and outer layers of the biofilm. *Nitrosomonas* play a crucial role in oxidizing ammonia to nitrite. However, it has been found that *Nitrospira* can perform complete nitrification. Therefore, it can be argued that *Nitrosomonas* and *Nitrobacter* act as the AOB and NOB, respectively, whereas *Nitrospira* plays a role in both the AOB and NOB [38]. A high ammonium loading rate and immediate substrate accessibility result in a higher relative abundance of *Nitrosomonas* and *Nitrospira* in the outer layer.

As shown in Table 4, the RBC bioreactor exhibited good COD removal efficiency throughout the experimentation period and achieved an average removal efficiency of 72.4 $\pm$ 2.5% with a 78.2 $\pm$ 7.5 mg/L effluent value, whereas the average effluent TN concentration was 1.54 $\pm$ 0.05 mg/L with a 38.3 $\pm$ 1.9% average removal efficiency. The RBC–ME bioreactor showed a further increase in the effluent removal efficiencies thanks to the incorporation of membrane filtration. A COD removal efficiency of 87.9 $\pm$ 3.2% with a 35 $\pm$ 8.9 mg/L effluent value and an average TN removal efficiency of 45.2 $\pm$ 0.7% with a 1.41 $\pm$ 0.05 mg/L average effluent value were obtained for RBC–ME.

Despite a low influent ammonium concentration, the RBC biofilm grew nitrifying bacteria, which significantly removed 95.8 $\pm$ 0.8% ammonium, while the RBC–ME maintained a higher ammonium removal efficiency of 98.9 $\pm$ 1.1%. The effluent ammonium concentration was as low as 0.03 $\pm$ 0.01 mg/L for both the RBC and RBC–ME, indicating a proficient nitrification process. An increase in the discharge nitrate value can be ascribed to the nitrifying bacteria activity [3].

Some reports have described that an RBC can undergo aerobic denitrification. A lower DO concentration at the bottom of an RBC facilitates denitrification [39]. However, a high C/N ratio and lower TN values restrict the nitrifying bacteria growth and thus, reduce the denitrification process. The system obtains a relatively lower TN removal because of lower influent quantities and strong competition between heterotrophic and autotrophic bacteria.

Higher removal efficiency for turbidity was achieved by the RBC–ME due to the membrane separation (see pore size in Table 3). The results show that the influent turbidity was 15.2 $\pm$ 0.6 NTU, which was significantly reduced to 3.3 $\pm$ 0.3 NTU and 0.32 $\pm$ 0.03 NTU in the RBC and RBC–ME, respectively, attributing to 78.9 $\pm$ 0.3% and 97.9 $\pm$ 0.1% removal efficiencies (Table 4). The effluent turbidity values for the RBC–ME are far better than the stand-alone RBC effluent. Thanks to low influent ammonium values, no substantial variations in the pH were detected during the experiments, and a neutral pH value was maintained.

Previous studies on the RBC bioreactor showed high organic and ammonium removal efficiency for both municipal and industrial wastewater [26,40]. In the self-refluxing RBC bioreactor for rural sewage treatment, a better treatment performance was obtained for a system with a 200% reflux ratio. The results show that the removal efficiency is more stable and better with reflux than without reflux. In the control with 0% reflux, the removal rates of COD, ammonium, and TN were 88.05 $\pm$ 3.17, 91.61 $\pm$ 3.26, and 41.58 $\pm$ 5.50%, respectively. Under 200% reflux, the removal rates of COD, ammonium, and TN improved, especially that of TN. For the 200% reflux ratio, the removal rates of COD, ammonium, and TN were up to 93.30 ± 7.35, 97.28 ± 5.94, and 74.21 ± 9.17%, respectively [18]. A non-woven RBC evaluated for the treatment of municipal wastewater supporting both aerobic and anaerobic processes resulted in a higher TN removal efficiency. Under the optimal conditions, the removal rates of COD and TN were 83.12% and 79.13%, respectively [41]. An RBC applied for the treatment of petroleum refinery wastewater resulted in 85.76% and 99.07% COD and ammonium removal, respectively [26]. The results suggested that an RBC may be considered as a promising method for petroleum refinery wastewater treatment, especially for simultaneous COD and ammonium removal.

3.4. Effect of Hydraulic Retention Time on COD Removal

Figure 4 shows the effect of HRT (12, 18, 24, and 48 h) on COD removal efficiency for both (RBC and RBC–ME) bioreactors. The results show that higher HRTs led to better COD removal efficiency in both bioreactors. A higher retention time means that microorganisms have a longer time to biodegrade the organics present in the wastewater [42]. As depicted in Figure 4, the maximum COD removal efficiencies of 80.9 ± 2.3% and 92.4 ± 2.4% were obtained for the RBC and the RBC–ME, respectively.

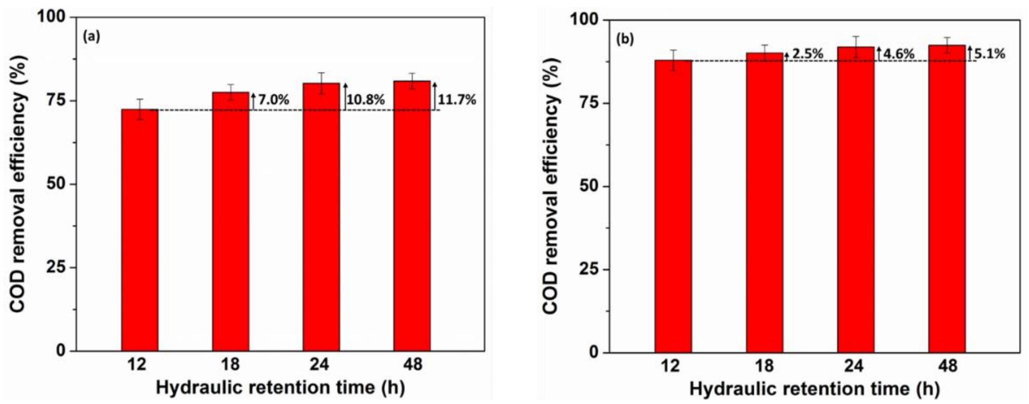


Figure 4. Effect of the HRT on the COD removal efficiency for the (**a**) RBC and (**b**) RBC–ME bioreactors.

In the RBC, COD removal efficiency increased from 72.4 ± 3.1% to 80.9 ± 2.3% as the HRT increased from 12 to 48 h. Increments of 7.0%, 10.8%, and 11.7% in COD removal efficiencies were observed for the HRTs of 18, 24, and 48 h, compared to 12 h (Figure 4a). On the other hand, RBC–ME exhibited an increase in COD removal efficiency from 87.9 ± 3.1% to 92.4 ± 2.4% as the HRT increased from 12 to 48 h. As shown in Figure 4b, an increase of 2.5%, 4.6%, and 5.1% in COD removal efficiency was observed for the HRTs of 18, 24, and 48 h, compared to 12 h. A higher COD removal efficiency confirms the effectiveness of membrane integration with an RBC. Such an advantage can be attributed to the presence of the biofilm on the membrane surface that further degrades the organics when the feed passes through it. The bioreactors perform well under higher HRTs; however, the HRT becomes limiting as it is directly related to the overall treatment capacity. Hence, careful selection of the optimum HRT becomes an essential part of the RBC design.

3.5. Effect of Hydraulic Retention Time on TN Removal

Figure 5 depicts the effect of TN removal efficiency at different HRTs (12, 18, 24, and 48 h) for both (RBC and RBC–ME) bioreactors. The results show that higher HRTs led to an increase in TN removal efficiency in both bioreactors. A higher retention time means that nitrifying microorganisms have prolonged contact time to biodegrade the nitrogen compounds present in the wastewater. Maximum TN removal efficiencies of 41.5 ± 0.8% and 48.6 ± 1.3% were obtained at a 48 h HRT for the RBC and RBC–ME, respectively.

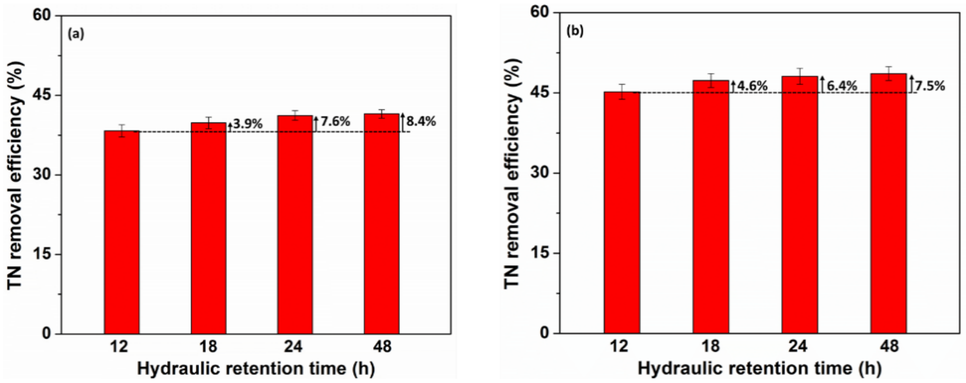


Figure 5. Effect of the HRT on the TN removal efficiency for the (**a**) RBC and (**b**) RBC–ME bioreactors.

In the RBC, TN removal efficiencies of 38.3 ± 1.2%, 39.8 ± 1.1%, 41.2 ± 0.9%, and 41.5 ± 0.8% were observed at the HRTs of 12, 18, 24 and 48 h, respectively. The RBC–ME exhibited an increase in TN removal efficiency from 45.2 ± 1.4% to 48.6 ± 1.3% as the HRT increased from 12 to 48 h (Figure 5). As shown in Figure 5b, increments of 4.6%, 6.4%, and 7.5% in TN removal efficiency were observed for the HRTs of 18, 24, and 48 h, compared to 12 h. As the HRT increases, the wastewater is retained longer in the bioreactor, allowing the microorganisms to biodegrade more nutrients. The results agree with the previous research that showed a long HRT led to a better degradation performance of municipal and industrial wastewater, recalcitrant pharmaceuticals, and micro-pollutants [43,44]. Ghalehkhondabi et al. [26] studied the performance of a four-stage RBC bioreactor for a petroleum refinery wastewater treatment. The increase in the HRT and reduction in HLR resulted in an increase in the organic and ammonium removal, and the maximum removal efficiencies of COD and ammonium obtained were 85.76% and 99.07%, respectively. In principle, a longer HRT is ideal for complete nitrification and high-strength wastewater treatment. Nevertheless, if the HRT of the reactor operation is shortened for operational or economic reasons, the influent concentration and microbial community could play a compensatory role in the biodegradation performance.

3.6. Effect of Hydraulic Retention Time on Turbidity

Figure 6 depicts the effect of turbidity removal efficiency at different HRTs (12, 18, 24, and 48 h) for both (RBC and RBC–ME) bioreactors. The results show that higher HRTs led to an increase in turbidity removal efficiency in both bioreactors. The maximum turbidity removal efficiencies of 84.2 ± 0.8% and 98.6 ± 1.3% were obtained at 48 h HRT for the RBC and RBC–ME, respectively. As the HRT increases, the wastewater is retained longer in the bioreactor, allowing the microorganisms to digest the suspended solid or allowing the solids to settle in the bioreactor. In the RBC, the turbidity removal efficiency increased from 78.9 ± 0.3% to 84.2 ± 0.8% as the HRT increased from 12 to 48 h. On the other hand, the RBC–ME exhibited a rise in turbidity removal efficiency from 97.9 ± 0.1% to 98.6 ± 1.3% as the HRT increased from 12 to 48 h. A higher turbidity removal efficiency confirms the effectiveness of membrane integration with an RBC. The membrane intercepts all the solids that otherwise contribute to the turbidity.

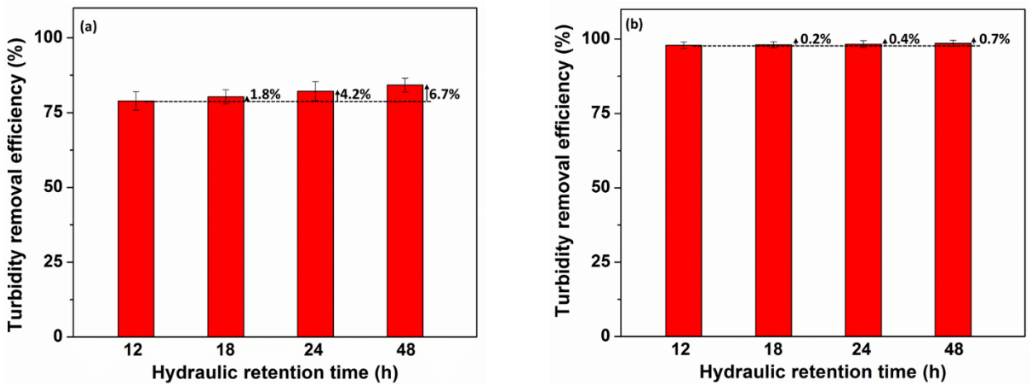


Figure 6. Effect of HRT on the turbidity removal efficiency for; (**a**) RBC and (**b**) RBC–ME bioreactors.

3.7. Membrane Permeability versus Hydraulic Retention Time in RBC–ME

Figure 7 shows the membrane permeability for short-term filtration in the RBC–ME bioreactor configuration. Membrane fouling is inevitable in almost all membrane processes [40,45]. This implies that a decline in permeability corroborates membrane fouling as a function of filtration time. As shown in Figure 7, the membrane permeability decreases sharply at the start of filtration, mainly owing to membrane pore blocking and irreversible adsorption of foulant. After that, permeability decreases steadily due to the slower rate of the deposition of foulant at the membrane surface, indicating reversible fouling. After reaching monolayer foulant adsorption, the affinity of foulant toward the membrane surface seems to be weaker as well. Toward the end of the filtration cycle, steady-state permeability is attained, which is ascribed to the development of a cake layer categorized as reversible fouling on a membrane that requires physical cleaning of the membrane [46,47]. The general trend is similar to the filtration of all parameters. A small rate of permeability decrease was still observed at the end of the filtration test, which is attributed to the buildup of foulant materials on the membrane surface. This occurred because no means of membrane fouling control was applied for the membrane filtration, an issue that can be addressed in a follow-up study. The current study was focused on the relative membrane fouling propensity of the RBC effluent under different operational HRTs.

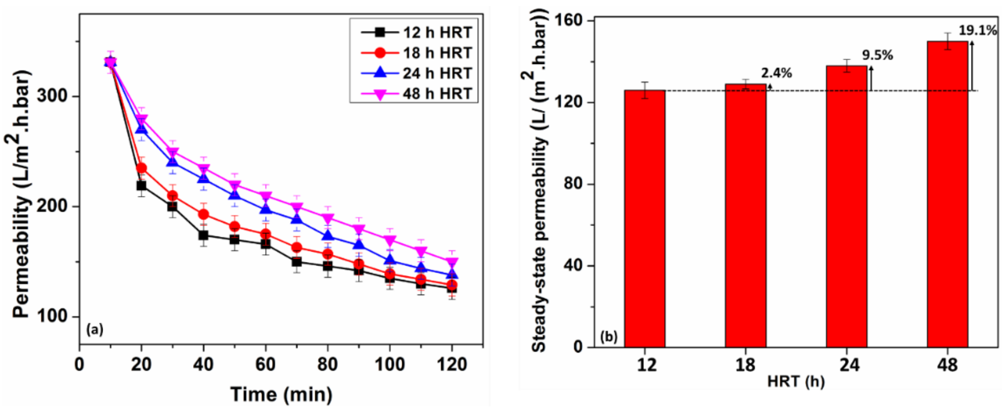


Figure 7. Evolution of the permeability as a function of filtration time at different HRTs for the RBC–ME configuration (**left**) and the summary of the steady-state permeabilities (**right**).

Figure 7 reveals the outcome of different HRTs on membrane permeability for the RBC–ME configuration. The steady-state permeabilities of 126, 129, 138, and 150 L/(m^2 h bar) were respectively attained at HRTs of 12, 18, 24, and 48 h, corresponding to filtration fluxes of 12.6, 12.9, 13.8, and 15.0 L/(m^2 h). Higher membrane permeability at an HRT of 48 h shows higher efficiency of organic compounds and suspended solids removal. This, in turn, reduces the membrane fouling and could potentially lower operating costs. The steady-state permeability increments of 2.4%, 9.5%, and 19.1% were obtained at 18, 24, and 48 h, respectively, compared to the 12 h HRT. This finding means that a higher HRT benefits from microbial degradation activity and nutrient removals coupled with better effluent filterability. Reduced membrane fouling mainly arose from the increase in the HRT and the reduction in the concentration polarization nearby the membrane surface due to a lower hydraulic loading rate in accordance with previous studies [13].

Reversible membrane fouling can be effectively controlled through optimizing parameters. In this study, the application of different HRTs has proven to be highly effective for membrane fouling control. Optimizing the parameters not only dampens the membrane fouling but also increases the effluent quality. The enhanced HRT significantly alleviated membrane fouling potential. The RBC–ME experienced severe fouling, as no fouling control technique was applied. Nevertheless, it is worth mentioning the trade-off of a high HRT and membrane fouling control that needs to be managed. A high HRT leads to a higher bioreactor volume and hence, higher investment costs. On the other hand, low membrane fouling propensity at high HRTs leads to lower costs associated with membrane fouling control as well as a lower membrane investment cost if the filtration is run at higher fluxes.

3.8. Membrane Fouling Analysis

Figure 8 shows the fouling resistance distribution in the RBC–ME bioreactor at different HRTs. The filtration resistances were measured at the end of each experiment. With an increase in the HRT from 12 to 18 h, the total fouling resistance (R_T) decreased from 3.25×10^{12} to 3.18×10^{12} m^{-1}, which further decreased to 2.97×10^{12} and 2.73×10^{12} m^{-1} at HRTs of 24 and 48 h, respectively. Since a constant membrane resistance (R_m) value of 4.55×10^{11} m^{-1} was obtained for all HRTs, both the pore blocking resistance and cake layer resistance played a crucial role in the total membrane fouling. The highest cake layer resistance (R_c) of 1.61×10^{12} m^{-1} at a 12 h HRT consisted of 49.6% of R_T, which decreased to 1.54×10^{12} m^{-1} at an HRT of 18 h, accounting for 48.6% of R_T. The R_c further decreased to 1.33×10^{12} m^{-1} and 1.09×10^{12} m^{-1} at HRTs of 24 and 48 h, respectively, consisting of 44.8% and 40% of R_T.

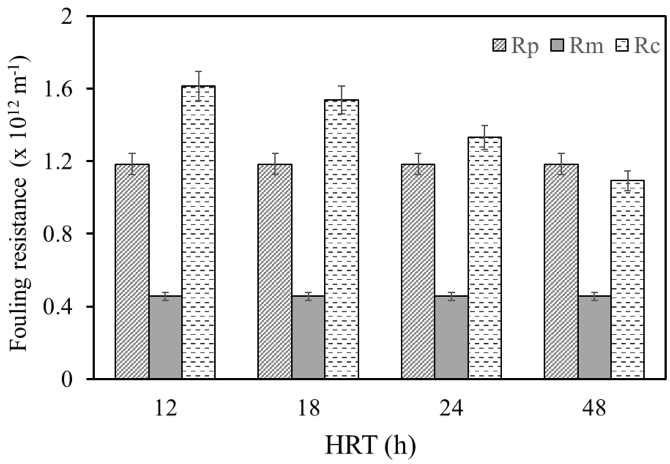


Figure 8. Fouling resistance distribution in the RBC–ME bioreactor at different HRTs.

A shorter HRT leads to insufficient degradation time for the organics and nutrients and results in the generation of more sludge, thereby increasing filtration resistance at shorter HRTs. Shorter HRTs also result in a higher secretion of extracellular polymeric substances and soluble microbial products, hence promoting membrane fouling [48,49]. The results of the membrane fouling analysis emphasized the importance of the HRT and recommended that the decline in membrane permeability at higher HRTs causes less membrane fouling and hence, increases the membrane permeability.

4. Conclusions

This study reports a high-performance membrane integrated with an RBC bioreactor as an efficient wastewater treatment process. The aim of the present research was to substitute the suspended growth system with an attached growth system. Therefore, the RBC, as an attached growth system, was coupled with an external UF membrane to treat synthetic domestic wastewater. The attached growth bioreactor creates the biofilm on the support media that provides better treatment efficiency than the suspended growth bioreactor due to the accumulation of a high microbial population over a large surface area. Therefore, better performance can be achieved by combining such a biofilm reactor as an RBC with a membrane, compared to suspended growth bioreactors. An increase in the HRT not only results in enhanced biological performance but also improves membrane permeability. The results show that the RBC exhibited 72.4 ± 2.5% COD, 38.3 ± 1.9% TN, 95.6 ± 0.8% ammonium, and 78.9 ± 0.3% turbidity removal efficiencies, while the RBC–ME showed better performance, with 87.9 ± 3.2% COD, 45.2 ± 0.7% TN, 98.9 ± 1.1% ammonium, and 97.9 ± 0.1% turbidity removal efficiencies. The highest COD and TN removal efficiencies were 92.4 ± 2.4% and 48.6 ± 1.3%, respectively, for the RBC–ME at an HRT of 48 h. The HRT enhancements resulted in 19.1% higher permeability at 48 h compared to 12 h. The biological and filtration performance of the RBC–ME reveals the economic impact and opens a great opportunity for significant improvements to the current membrane technology. A membrane-integrated RBC poses an attractive alternative to treat wastewater in the decentralized and open-air canal systems where the problem of a large footprint is less important.

Author Contributions: S.W. performed all the experiments and prepared the manuscript draft; M.R.B. and N.Y.H. contributed to the original idea of the study, the conceptual design of the study, supervised the work, and revised the manuscript; N.A.H.M.N., N.H., J.R., N.S., Y.W. and A.L.K. revised the manuscript. All authors have read and agreed to the published version of the manuscript.

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