ORIGINAL PAPER



Fouling behaviour of industrial waste-based ceramic membrane in anaerobic membrane bioreactor treating low strength wastewater

S. Dhiman¹ · S. Yadav² · M. Balakrishnan² · N. Ahsan¹

Received: 22 June 2022 / Revised: 29 March 2023 / Accepted: 17 June 2023 / Published online: 28 June 2023 © The Author(s) under exclusive licence to Iranian Society of Environmentalists (IRSEN) and Science and Research Branch, Islamic Azad University 2023

Abstract

This work describes the fouling of industrial waste sugarcane bagasse ash ceramic membrane (pore size ~ 8.6 µm; water permeability 27.2×10^3 L/m² h bar) in wastewater treatment in anaerobic membrane bioreactor (AnMBR). AnMBR system was operated in sequential batch reactor (SBR) mode at 18 h hydraulic retention time for 31 days. For influent concentration of 171 ± 12 mg COD/L, average chemical oxygen demand (COD) removal was high (~94%). Biomass activity of anaerobic sludge improved from 0.15 (day 1) to 0.35 mg COD_{removed}/mg MLVSS.d (day 31). Operating flux was maintained at 17.8 ± 1.4 L/m² h and the transmembrane pressure reached up to 170 mbar on day 31, increasing at a rate of 15.7 mbar/d. Specific bound extracellular polymeric substance (EPS) concentration was much higher in surface deposits (356 mg EPS/g MLVSS) than in the reactor sludge suspension (32.3 ± 14.4 mg EPS/g MLVSS). Though SBR is a good alternative configuration to reduce membrane surface) still cannot be prevented. Calculation of total filtration resistance (R_t) showed the resistance of the caked surface deposits, R_c (2.19×10^{12} m⁻¹) to be dominant at 83% of R_t. Of the two fouling control strategies tested viz. filtration-relaxation (4 min–1 min) and permeate backflushing (up to 3 times operating flux), backflushing was more effective. These findings indicate the potential of these alternative membranes in wastewater treatment application; at the same time, further investigations are required to minimize membrane fouling.

Keywords Sugarcane bagasse ash ceramic membrane \cdot Anaerobic membrane bioreactor \cdot Sequential batch reactor \cdot Low strength wastewater \cdot Membrane surface deposits \cdot Extracellular polymeric substances

Introduction

Ceramic membranes (CMs) are increasingly being investigated in wastewater treatment applications (Hofs et al. 2011; Zielińska et al. 2016; Hubadillah et al. 2020; Delikanli et al. 2022) especially for filtration of industrial effluents such as oil-in-water (Rasouli et al. 2019; Liu et al. 2020; Wang et al. 2021) and for sludge separation in aerobic/anaerobic membrane bioreactors (Ae/AnMBR) (Hasan et al. 2011;

Editorial responsibility: Samareh Mirkia.

S. Dhiman sourbh67@gmail.com

Yue et al. 2015; Aslam et al. 2018; Nilusha et al. 2020; Chen et al. 2021). The global CM industry with a valuation of \$10,893 million is projected to have a compound annual growth rate of 11.3% over the period 2019-2027 (Arumugham et al. 2021). CMs display higher stability to chemical and thermal changes (Abdullayev et al. 2019); they also show higher fouling resistance compared to polymeric membranes (Hofs et al. 2011; Chen et al. 2021). The cost of conventional raw materials (e.g., alumina, zirconia, TiO₂) for fabricating CMs is however high, so the use of CMs tends to be somewhat limited (Abdullayev et al. 2019). To counter this problem, CMs prepared with low-cost (waste) raw materials has gained interest in recent years. Such alternative materials include, for instance, cow bones (Hubadillah et al. 2020), sugarcane bagasse ash (SBA) (Jamalludin et al. 2018), rice husk ash (Lawal et al. 2020), coal fly ash (Ahmad et al. 2021; Wang et al. 2021), clay (Shafiquzzaman et al. 2020) and mixture of various silicates (Zuriaga et al. 2017). CMs manufactured from waste raw materials have



¹ Department of Civil Engineering, Faculty of Engineering and Technology, Jamia Millia Islamia, New Delhi 110025, India

² The Energy and Resources Institute (TERI), Darbari Seth Block, IHC Complex, Lodhi Road, New Delhi 110003, India

the potential to be viable at industrial scale. For instance, the REMEB (REcycled MEmbrane Bioreactor) project demonstrated the use of a combination of wastes (olive stones, chamotte, marble powder) for low-cost CM manufacturing, with the performance of this 2 µm pore size membrane being validated at industrial scale at the Aledo wastewater treatment plant in Spain (Zuriaga et al. 2017). Another pilot involving membranes fabricated from waste pyrophyllite and alumina was tested in a water resource recovery facility in S Korea (Jeong et al. 2017b). Among low-cost materials for CM preparation, the most common are clays (29%), kaolin (27%) and coal fly ash (17%) with only 4% based on SBA (Abdullayev et al. 2019). The annual global production of SBA is 12.6 million metric tonnes (Neto et al. 2021). It has high content of silica (65% by weight) with traces of oxides of aluminium (0.49%), titanium (0.08%), iron (0.49%), calcium (2.75%), magnesium (3.26%) (Umamaheswaran and Batra 2008); also, there are silanols group on the surface (Jamalludin et al. 2018). These characteristics makes the material eligible for separation, catalysis and adsorbent processes, and also imparts strength, making SBA a suitable choice as raw material for CM production.

Towards an effort to develop sustainable options for wastewater treatment, waste-based CMs can be combined with anaerobic treatment to exploit the benefits of low energy requirements and low sludge yield. One of the primary challenges in such AnMBRs is membrane fouling from soluble microbial products (SMP) and extracellular polymeric substances (EPS) (Chen et al. 2022). Composition of EPS i.e. concentration of proteins, carbohydrates and fraction of bound EPS plays a major role in determining the severity of fouling (Jang et al. 2021). High protein content is likely to enhance cake fouling due to accumulation of flocculated larger particles on the membrane surface with less pore blockage (Liu et al. 2021). High concentration of tightly bound EPS (TB-EPS) in the sludge deposits also increases fouling (Liu et al. 2012). Properties of the deposits influence the fouling mechanism with microparticles $(5-10 \ \mu m)$ contributing to cake layer formation while colloidal (0.45-1 µm) and sub-micrometer (1-5 µm) particles contribute to the pore blockage phenomenon (De Vela 2021). The significance of membrane fouling has led to the development of multiple fouling control methods in AnMBRs viz. (a) filtration-relaxation (Chu et al. 2005; Giménez et al. 2011; Huang et al. 2011; Lin et al. 2011; Robles et al. 2013; Chen et al. 2017; Aslam et al. 2018; Liu et al. 2018; Ji et al. 2020) (b) permeate backflushing (Chu et al. 2005; Ho et al. 2007; Giménez et al. 2011; Robles et al. 2013; Smith et al. 2013; Aslam et al. 2018) (c) gas sparging (Smith et al. 2013; Chen et al. 2017) (d) physical/chemical cleaning (Chu et al. 2005; Liu et al. 2012, 2018; Chen et al. 2017) and (e) use of additives e.g. powdered or granular activated carbon (Chen

🕈 🛛 🖉 Springer

et al. 2021; Balcioğlu et al. 2022). Yet another approach is to modify the operation to minimize membrane exposure to sludge and thereby reduce fouling. One such option is the sequential batch reactor (SBR) configuration wherein the settled bioreactor effluent is subjected to filtration. Compared to conventional MBRs, the transmembrane pressure (TMP) build-up is more effectively controlled in SBR–MBRs (Zhang et al. 2006).

There is extensive literature on the development, characterization and lab-scale testing of waste-based CMs for wastewater treatment (Balakrishnan et al. 2020; Goswami et al. 2022; Hubadillah et al. 2022). Fouling behavior for such membranes needs to be better understood considering the complex interplay among the membrane material characteristics, wastewater properties and operation parameters, especially in MBR applications where the biological performance has a direct impact on the membrane filtration (Wu and Lee 2011; Robles et al. 2013). Though waste-based CMs have been tested in MBRs (e.g., Hasan et al. 2011; Lawal et al. 2020), very few studies have examined fouling mitigation strategies. Basu et al. (2014) used SBA based CM in baffled aerobic-anoxic MBR treating nitrate-rich water wherein biopolymer foulants (EPS and SMP) were discussed but not the approach to control fouling. In another study employing pyrophyllite wastebased CM in domestic wastewater treatment in AnMBR, gas sparging was employed for fouling control (Jeong et al. 2017a). The TMP remained low (0.03 bar until 76 days) but the corresponding operational flux at 18 h hydraulic retention time (HRT) was also very low $(2.7 \text{ L/m}^2 \text{ h})$.

In an earlier study with SBR-AnMBR system using SBA based membrane, the effect of varying HRTs and feast-famine cycles on system performance including membrane fouling was investigated (Dhiman et al. 2023). This work aims to further understand fouling behavior and its control in an AnMBR equipped with waste SBA based membrane operating at optimal HRT. Membrane fouling due to EPS and the effectiveness of two fouling control strategies-filtration-relaxation and permeate backflushing were investigated. The following aspects are unique to this study: (a) SBA based CM (b) realistic permeate flux, considering the maximum reported flux of 16–18 L/m² h for pilot/full scale AnMBR systems treating municipal wastewater (Chen et al. 2021; Kong et al. 2021) (c) low strength wastewater feed (representative of urban drains) and (d) SBR mode of operation. To the best of our knowledge, no study has been reported on the performance of SBA based CM in AnMBR under these conditions. The findings are expected to contribute towards optimizing the integrated biological-filtration process in this AnMBR to enable long-term operation with reduced fouling. This study was conducted in The Energy and Resources Institute (TERI), New Delhi, India in 2021.

Materials and methods

AnMBR set-up and operation

Figure 1 shows the experimental set-up of AnMBR operated in SBR mode.

The AnMBR set-up consisted of three units: continuous stirred tank reactor (CSTR) maintained under anaerobic conditions, holding tank containing the CSTR effluent and filtration tank housing the membrane module. All units were of transparent polymethyl methacrylate (acrylic) and were fabricated locally. The CSTR working volume was 6.5 L; only 5 L permeate was filtered in each HRT cycle. Operation was conducted in SBR mode and the CSTR was fed using reverse osmosis (RO) booster pump (Kemflo, India). The different stages in SBR mode were: feeding (10 min), settling (1 h), reaction (16.5 h) and decantation (20 min). N₂ purging was done for 20 min after every feeding to ensure anaerobic environment in the CSTR; the temperature was monitored using digital thermometer (Aeoss, India). The CSTR effluent was collected in a holding tank and thereafter fed to the filtration tank equipped with submerged flat sheet CM composed of SBA. The membrane modules (pore size $8.6 \pm 1.4 \mu m$; effective filtration area 0.0233 m²) were prepared in-house. Inflow and outflow of the filtration tank were maintained using peristaltic pumps (Electrolab India Pvt. Ltd., India). TMP was measured using mercury manometer. HRT was maintained at 18 h based on reported work (Dhiman et al. 2023). Only one HRT cycle (18 h) was completed over 24 h and next cycle was started the following day; thus, the system was idle for 6 h in each cycle. Filtration-relaxation (4 min-1 min, throughout the operation) and permeate backflushing (backflushing/operating flux ratio i.e. J_b/J up to 3, once every HRT cycle when a drop in the flux was observed) were employed for fouling control. The system was operated for 6 days a week (from Monday to Saturday); the experimental facility was unavailable on Sunday following COVID restrictions.

Synthetic feed and seed sludge

Synthetic feed as per Shim et al. (2002) was modified to obtain low strength wastewater. This feed was prepared with tap water and had the following composition (concentration in mg/L): D-glucose (C₆H₁₂O₆) 100, L-glutamic acid $(C_5H_9NO_4)$ 50, ammonium acetate (CH_3COONH_4) 40, sodium hydrogen carbonate (NaHCO₃) 65, ammonium chloride (NH₄Cl) 35, potassium dihydrogen phosphate (KH₂PO₄) 28, sodium chloride (NaCl) 12, magnesium chloride hexahydrate (MgCl₂·6H₂O) 8, calcium chloride dihydrate (CaCl₂ \cdot 2H₂O) 6, ferric chloride hexahydrate (FeCl₃·6H₂O) 2. The feed chemical oxygen demand (COD) was~171 mg/L. Seed sludge, with mixed liquor volatile suspended solids (MLVSS) of 1174 mg/L, was taken from the same AnMBR system that previously treated low strength wastewater for a period of 1 year followed by a 5 month shut-down. For this study, initial MLVSS in CSTR was adjusted to $\sim 600 \text{ mg/L}$.

Analytical methods

Feed, CSTR effluent and permeate samples were analyzed for COD, ammonia, phosphate, pH and turbidity; COD, pH and turbidity were measured twice a week while all other parameters were measured once a week. The CSTR effluent was filtered through Whatman grade 1 filter to remove particulate matter prior to analysis. Mixed liquor suspended solids (MLSS) and MLVSS analysis were done weekly for CSTR sludge suspension, CSTR effluent and permeate. All analyses were done at least in duplicate as per APHA, 2005 protocols. The CSTR sludge suspension was analyzed for EPS fractions (soluble, loosely bound (LB) and TB) and for both proteins and carbohydrates (extracted using method provided by Li and Yang 2007). Proteins and carbohydrates were quantified using methods of Hatree (1972) and Dubois et al. (1956), respectively. Membrane surface deposits were collected at the end of operation by washing the membrane with 100 mL RO water and the suspension was analyzed for MLVSS and EPS.



Fig. 1 Illustration of AnMBR experimental set-up



To assess adaptation to operation conditions, sludge from the CSTR was subjected to biomass activity analysis. Sludge suspensions sampled on day 1 and day 31 were introduced in Schott bottles; the MLVSS concentration was kept constant for both samples. Tap water was used to adjust the final volume to 500 mL. Biomass activity assessment was done over 3 cycles: cycle 1 (72 h), cycle 2 (48 h) and cycle 3 (48 h). At the start of each cycle, N₂ purging was done for 10 min to provide oxygen free environment and then the sodium acetate substrate (594 mg/L)was fed. Samples were withdrawn at periodic intervals during each cycle to assess substrate removal in terms of COD reduction. MLVSS analysis was done at the start of cycle 1 and the end of cycle 3. Equation (1) was used to calculate the biomass activity of the sludge (Tomar et al. 2018).

$$R_t = R_{int} + R_c + R_p = \frac{TMP}{J \cdot \eta}$$
(3)

where R_t (m⁻¹) is total filtration resistance, R_{int} (m⁻¹) is membrane intrinsic resistance, R_c (m⁻¹) is resistance due to cake layer or surface deposits and R_p (m⁻¹) is the resistance experienced due to pore blockage. *TMP* (Pa) is the transmembrane pressure, J (m/s) is the permeate flux and η is the dynamic viscosity (Pa s) of the permeate (assumed to be of that water at ambient temperature).

 R_{int} was determined by filtering RO water through virgin membrane. R_t was determined by filtering RO water through the used membrane after AnMBR operation ended. Surface deposits were removed from membrane and RO water was filtered to obtain sum of R_p and R_{int} . R_c was calculated by subtracting R_p and R_{int} from R_t .

$$Activity\left(\frac{mg\ COD\ removed}{mg\ MLVSS\ day}\right) = Slope\ of\ graph\left(\frac{mg\ COD\ removed}{h}\right) * \frac{24\ h}{day} * \frac{1}{mg\ MLVSS}$$
(1)

Membrane characterization

Water permeability of the virgin membrane was determined using a locally fabricated acrylic tank (3 L) wherein the flat sheet CM module (5.5 cm × 5.5 cm) was submerged. The tank was fed with tap water manually and the permeate flow was maintained using peristaltic pump (Acuflo, Arrow Weighting System Pvt. Ltd., India). Flux was determined volumetrically with a measuring cylinder and stop watch. TMP was measured using mercury manometer. Water absorption of the virgin membrane was determined as per ISO:10,545 (International Organization for Standardization). The dry CM (12 cm × 12 cm) was weighed initially (W_A), immersed in boiling water for 2 h and then allowed to cool naturally for 4 h in the water. Excess surface water was removed by patting with slightly wet cloth and the CM was weighed again (W_B). Water absorption was calculated using Eq. (2).

Water absorption (%) =
$$\frac{(W_B - W_A)}{W_A} \times 100$$
 (2)

Porosimetry analysis of the virgin and fouled membrane was done using mercury porosimeter (Micromeritics, AutoPore IV, USA).

Membrane fouling study

Membrane filtration resistance (R_t) was calculated quantitatively by resistance-in-series model using Eq. (3) (Liu et al. 2012).

Results and discussion

Membrane characterization

Porosity, pore diameter and bulk density of the virgin CM were $46.06 \pm 2.01\%$, $8.62 \pm 1.40 \ \mu\text{m}$ and $1.31 \pm 0.05 \ \text{g/mL}$ respectively. Water absorption was in the range of 32-35%. There is a linear relation between water flux J_{tw} and TMP ($R^2 = 0.9965$; Supplementary sheet Fig. S1) for this micro-filtration membrane, following Darcy's law for permeable media (Qiu et al. 2010). The water permeability was $27.2 \times 10^3 \ \text{L/m}^2$ h bar. Water flux depends more on pore size compared to porosity (Liu et al. 2020). Against conventional CMs prepared from alumina and natural zeolites, the SBA CM has marginally higher pore size and water permeability (Table 1). The threshold gradient pressure (TGP) i.e. the minimum TMP required for permeate flow to occur (Yin et al. 2018) is identified to be 1.31 mbar (Supplementary sheet Fig. S1).

AnMBR performance

Organics and nutrients removal

Figure 2 shows the COD concentration and removal profile in AnMBR operated at a fixed HRT of 18 h.

For the initial 6 days, only biological treatment took place. Membrane filtration commenced on day 7 with a permeate COD of 2.2 mg/L and ~99% removal efficiency that was maintained over the following days. The system had to be shut-down for 10 days (day 12-21) due to mechanical malfunction in the filtration unit. Upon restarting,



 Table 1
 Comparison of CMs

 prepared from different raw
 materials

Average pore size (µm)	Porosity (%)	Permeability (L/ m ² h bar)	Reference
5.9	35	26.6×10^3	Dong et al. (2006)
2.13	n.s.*	22.6×10^{3}	Fang et al. (2011)
2.2	46	7.6×10^{3}	Qin et al. (2017)
4.6	31	17.9×10^{3}	Yin et al. (2018)
1–2	30	8.6×10^{3}	Zou et al. (2019)
0.92	46.7	0.7×10^{3}	Mouratib et al. (2020)
8.6	39.8	$27.2 \times 10^{3^{**}}$	Present work
	Average pore size (µm) 5.9 2.13 2.2 4.6 1-2 0.92 8.6	Average pore size (µm)Porosity (%)5.9352.13n.s.*2.2464.6311-2300.9246.78.639.8	Average pore size (µm)Porosity (%) m^2 h bar)Permeability (L/ m^2 h bar)5.935 26.6×10^3 2.13n.s.* 22.6×10^3 2.246 7.6×10^3 4.631 17.9×10^3 1-230 8.6×10^3 0.9246.7 0.7×10^3 8.639.8 $27.2 \times 10^{3**}$

*n.s.: not specified

**With tap water; all others used deionized water





Fig. 2 COD removal profile in AnMBR (18 h HRT)

the COD removal was > 90% with average removal of $93.6 \pm 3.6\%$ until day 31. MLVSS increased from 594 (day 1) to 1176 mg/L (day 31) with improved MLSS/MLVSS ratio 0.68. There was no ammonia removal in the CSTR but around 40% removal was observed in the permeate probably due to passive aeration in the holding tank. Phosphate removal in the permeate was around 80%.

Biomass activity results showed 0.15 mg $\text{COD}_{\text{removed}}$ / mg MLVSS.d on day 1 that increased to 0.35 mg $\text{COD}_{\text{removed}}$ / mg MLVSS.d on day 31 (Fig. 3). This indicates that the reactor sludge, fed on low strength synthetic wastewater (171 mg COD/L), could quickly adapt to change in substrate at a higher concentration (594 mg sodium acetate/L).

Filtration performance

Figure 4 shows the operating flux vs TMP profile during filtration in the AnMBR system.

Filtration was started on day 7 in filtration-relaxation (4 min–1 min) mode; no permeate backflushing was applied initially. When continuous flux drop was observed, permeate backflushing was employed as specified in Fig. 4. The system was shut-down for 10 days (day 12–21) due to system malfunction as mentioned earlier. On day 22, filtration

Fig. 3 Biomass activity of sludge sample on day 1 and day 31: cumulative COD removal for cycle 3 (48 h); COD removal stabilized within 8 h for both samples

was commenced but without backflushing. Flux equal to the desired value was recorded without any backflushing probably due to dislodging of loose surface deposits from the membrane surface during the shut-down period. Subsequently, the flux dropped by 8% on day 23. To maintain constant operating flux, backflushing was employed again but the flux could not be recovered fully. Figure 5 shows the TMP experienced during backflushing, with TMPi being the value when backflushing was initiated (t=0 min) and TMPf when the backflushing was terminated (t = 4 min). When $J_{\rm b}/J$ ratio was 3 (day 28-31), at the conclusion of every backflush cycle, a TMP of ~39 mbar was recorded indicating residual fouling which could not be removed. Operation was terminated on day 31 with average flux for the operating period at 17.8 ± 1.43 L/m² h (8% less than desired flux) and TMP increase at the rate of 15.7 mbar/d. Quality of permeate obtained was good with very low solids concentration $(4.8 \pm 5.6 \text{ mg/L})$ with up to 99% solids removal. Turbidity of the permeate was also correspondingly low $(1.5 \pm 0.4 \text{ NTU})$.

In this work, relatively high flux was achievable (to the best of our knowledge) compared to past studies using waste-based CMs in AnMBR. Hasan et al. (2011) studied rice bran and clay-based CM in MBR configured in



Backflush TMP (mbar)

Fig. 4 Flux and TMP variation profile in AnMBR (18 h HRT). i flux dropped by 20% and permeate backflushing was applied subsequently as per the following ratios **ii** $J_b/J = 2$, **iii** $J_b/J = 3$,



22 23 24 25 28 29 30

(iv) (v) 200

180

160

140

120

100

80 60 40

20

0

FMP (mbar)

0

0



Fig. 5 Effect of backflush/operating flux ratio on fouling

■LB protein ■TB protein ■LB carbohydrates ■TB carbohydrates ■Soluble EPS

aeration, intermittent aeration and non-aeration modes with corresponding fluxes of $4.1-8.3 \text{ L/m}^2$ h, 3.04 L/m² h and 0.875 L/m² h, respectively. In another AeMBR study, rice husk ash and clay-based CM showed a flux of 4.5 L/m² h (Lawal et al. 2020). In lab-scale AnMBR and AeMBR studies employing waste pyrophyllite based CMs, the flux was $1.1-2.7 \text{ L/m}^2$ h (Jeong et al. 2017a) and 5 L/ m^2 h, respectively (Jeong et al. 2017b). Though a higher flux of 15 L/m^2 h was maintained in pilot operations with AeMBR system, sharp TMP rise to 0.3 bar was encountered within 10 days (Jeong et al. 2017b).

Operation in the SBR mode, with a separate holding tank for the decanted bioreactor effluent, helped retain bulk of the sludge in the bioreactor. However, fine sludge particles did not settle and eventually deposited on the membrane surface in the filtration unit. Cake formation could not therefore be completely prevented. In conventional operation integrating biological treatment and membrane filtration in a single reactor (data not shown), TMP build-up rate was high (i.e. 44 mbar/d corresponding to a flux of 9.6 L/m^2 h) and physical cleaning of the membrane was required within 3 days of commencing operation. In Fig. 6 Soluble and specific bound EPS profiles of the CSTR sludge suspension in AnMBR system

contrast, the SBR mode of operation showed comparatively slower TMP build-up.

Membrane fouling

Figure 6 shows the EPS profiles of the CSTR sludge suspension in the AnMBR system.

The EPS constituents viz. proteins and carbohydrates are non-settleable organic components which can affect biofilm formation and hence fouling (Robles et al. 2013; Zhao et al. 2014; Xiong et al. 2016). Both proteins and carbohydrates were analyzed in the soluble, LB and TB fractions. Specific bound EPS (i.e. mg EPS/g MLVSS) in the sludge suspension ranged from 16 to 48 mg/g MLVSS with an average value of 34.9 ± 13.6 mg/g MLVSS for the operation period. Average soluble EPS was 11.1 ± 3.0 mg/L. After operation, surface deposits were analyzed for MLVSS and EPS. MLVSS was 300 mg/L and specific bound EPS was found to be 356.6 mg/g MLVSS (proteins: 298.4 mg/g MLVSS, carbohydrates: 58.2 mg/g MLVSS), approx. 10 times higher than in the sludge suspension. This could be the cause of fouling as high EPS can result in accumulation of particles



Fig. 7 Membrane fouling resistance distribution



Fig. 8 Shift in pore diameter of used (fouled) membrane

on membrane surface increasing resistance to permeate flow (Chen et al. 2017; Aslam et al. 2018; Nilusha et al. 2020). TB-EPS concentration was high too in surface deposits i.e. 212 mg/g MLVSS compared to LB-EPS (144.6 mg/g MLVSS) which is not easily removable because of tight bond with the membrane (Liu et al. 2012). Figure 7 shows the total membrane resistance at the end of the 31-day AnMBR operation.

The highest resistance contribution is by surface deposits $(2.19 \times 10^{12} \text{ m}^{-1}, 83\%)$, followed by the intrinsic $(3.10 \times 10^{11} \text{ m}^{-1}, 12\%)$ and pore-blockage resistance $(1.35 \times 10^{11} \text{ m}^{-1}, 5\%)$. Much higher fouling resistance due to surface deposits may be attributed to rapid surface deposition rate even before membrane pore saturation; this highlights the need for further optimization of fouling control methods to minimize cake layer formation for this membrane. Porosity and pore diameter of the used (fouled) membrane were $39.78 \pm 1.40\%$ and $6.63 \pm 0.57 \mu m$ respectively. Bulk density increase (to $1.41 \pm 0.02 \text{ g/mL}$) and shift in peak in the log differential intrusion vs pore size plot (Fig. 8) were also observed. These changes were attributed to the marginal pore blockage of the membrane during operation.

Conclusion

AnMBR equipped with waste SBA based CM was studied to treat low strength wastewater in SBR mode. A fixed HRT of 18 h was adequate for high COD removal of ~94%. SBR is a good alternative configuration to reduce membrane exposure to sludge and hence control the fouling rate; however, formation of cake layer (due to deposition of sludge fines on the membrane surface) cannot be prevented. Membrane surface deposits with high EPS concentration is the key contributor to fouling; backflushing is required to dislodge the deposits. Further investigations are required to minimize the surface deposits and thus control the fouling with this waste-based CM.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s13762-023-05070-w.

Acknowledgements The authors are thankful to TERI for the research facility to conduct this work. The membrane mercury porosimetry analysis done by R. K. Singh, TERI is also greatly appreciated. The constructive comments provided by the anonymous reviewer that helped us to improve the manuscript are much appreciated.

Authors contributions SD: Conducting AnMBR experiments, data analysis, writing. SY: Membrane preparation and characterization, data analysis. MB: Supervision, review and editing. NA: Supervision.

Funding S. Dhiman is grateful to University Grants Commission for Junior and Senior Research Fellowship (UGC Ref. No. 3781/NET-DEC2018).

Availability of data and materials Not applicable.

Declarations

Conflict of interest The authors declare no conflict of interests.

Ethical approval No applicable.

Consent to participate Not applicable.

Consent to publication All co-authors agree to the publishing of this work in International Journal of Environmental Science and Technology.

References

- Abdullayev A, Bekheet MF, Hanaor DA, Gurlo A (2019) Materials and applications for low-cost ceramic membranes. Membranes 9(9):105. https://doi.org/10.3390/membranes9090105
- Ahmad B, Dilshad MR, Haider B, Anwar MM, Ali H, Gilani SMA, Ahmad HB, Farooq M (2021) Synthesis of novel fly ash based geo-polymeric membranes for the treatment of textile waste water. Int J Environ Sci Technol. https://doi.org/10.1007/ s13762-021-03527-4
- Arumugham T, Kaleekkal NJ, Gopal S, Nambikkattu J, Rambabu K, Aboulella AM, Wickramasinghe SR, Banat F (2021) Recent developments in porous ceramic membranes for wastewater treatment



and desalination: a review. J Environ Manag 293:112925. https:// doi.org/10.1016/j.jenvman.2021.112925

- Aslam M, Yang P, Lee PH, Kim J (2018) Novel staged anaerobic fluidized bed ceramic membrane bioreactor: energy reduction, fouling control and microbial characterization. J Membr Sci 553:200–208. https://doi.org/10.1016/j.memsci.2018.02.038
- Balakrishnan M, Batra VS, Dikshit PK, Yadav S (2020) Silica MBRs: design, configuration, and performance. Current trends and future developments on (bio-) membranes. Elsevier, pp 31–91. https:// doi.org/10.1016/B978-0-12-816822-6.00002-1
- Balcıoğlu G, Vergili I, Gönder ZB, Yilmaz G, Bacaksiz AM, Kaya Y (2022) Effect of powdered activated carbon addition on membrane performance and fouling in anaerobic membrane bioreactor. Int J Environ Sci Technol. https://doi.org/10.1007/s13762-022-04203-x
- Basu S, Singh SK, Tewari PK, Batra VS, Balakrishnan M (2014) Treatment of nitrate-rich water in a baffled membrane bioreactor (BMBR) employing waste derived materials. J Environ Manag 146:16–21. https://doi.org/10.1016/j.jenvman.2014.07.022
- Chen R, Nie Y, Hu Y, Miao R, Utashiro T, Li Q, Xu M, Li YY (2017) Fouling behaviour of soluble microbial products and extracellular polymeric substances in a submerged anaerobic membrane bioreactor treating low-strength wastewater at room temperature. J Membr Sci 531:1–9. https://doi.org/10.1016/j.memsci.2017.02. 046
- Chen C, Sun M, Liu Z, Zhang J, Xiao K, Zhang X, Song G, Chang J, Liu G, Wang H, Huang X (2021) Robustness of granular activated carbon-synergized anaerobic membrane bioreactor for pilot-scale application over a wide seasonal temperature change. Water Res 189:116552. https://doi.org/10.1016/j.watres.2020.116552
- Chen C, Sun M, Chang J, Liu Z, Zhu X, Xiao K, Song G, Wang H, Liu G, Huang X (2022) Unravelling temperature-dependent fouling mechanism in a pilot-scale anaerobic membrane bioreactor via statistical modelling. J Membr Sci 644:120145. https://doi.org/ 10.1016/j.memsci.2021.120145
- Chu LB, Yang FL, Zhang XW (2005) Anaerobic treatment of domestic wastewater in a membrane-coupled expended granular sludge bed (EGSB) reactor under moderate to low temperature. Process Biochem 40(3–4):1063–1070. https://doi.org/10.1016/j.procbio. 2004.03.010
- De Vela RJ (2021) A review of the factors affecting the performance of anaerobic membrane bioreactor and strategies to control membrane fouling. Rev Environ Sci Biotechnol 20(3):607–644. https:// doi.org/10.1007/s11157-021-09580-2
- Delikanli NE, Harman BI, Yigit NO, Sardohan Koseoglu T, Kitis M, Koseoglu H (2022) Separation of Cu²⁺ from membrane concentrate by SiO₂/3-aminopropyltriethoxysilane-coated ceramic membrane. Int J Environ Sci Technol 19(1):379–390. https://doi.org/ 10.1007/s13762-021-03157-w
- Dhiman S, Balakrishnan M, Naddeo V, Ahsan N (2023) Performance of anaerobic membrane bioreactor (AnMBR) with sugarcane bagasse ash-based ceramic membrane treating simulated lowstrength municipal wastewater: effect of operation conditions. Water Air Soil Pollut 234(3):141. https://doi.org/10.1007/ s11270-023-06173-3
- Dong Y, Chen S, Zhang X, Yang J, Liu X, Meng G (2006) Fabrication and characterization of low cost tubular mineral-based ceramic membranes for micro-filtration from natural zeolite. J Membr Sci 281(1–2):592–599. https://doi.org/10.1016/j.memsci.2006.04.029
- Dubois M, Gilles KA, Hamilton JK, Rebers PT, Smith F (1956) Colorimetric method for determination of sugars and related substances. Anal Chem 28(3):350–356. https://doi.org/10.1021/ac60111a017
- Fang J, Qin G, Wei W, Zhao X (2011) Preparation and characterization of tubular supported ceramic microfiltration membranes from fly

ash. Sep Purif Technol 80(3):585–591. https://doi.org/10.1016/j. seppur.2011.06.014

- Giménez JB, Robles A, Carretero L, Durán F, Ruano MV, Gatti MN, Ribes J, Ferrer J, Seco A (2011) Experimental study of the anaerobic urban wastewater treatment in a submerged hollow-fibre membrane bioreactor at pilot scale. Bioresour Technol 102(19):8799– 8806. https://doi.org/10.1016/j.biortech.2011.07.014
- Goswami KP, Pakshirajan K, Pugazhenthi G (2022) Process intensification through waste fly ash conversion and application as ceramic membranes: a review. Sci Total Environ 808:151968. https://doi. org/10.1016/j.scitotenv.2021.151968
- Hasan MM, Shafiquzzaman M, Azam MS, Nakajima J (2011) Application of a simple ceramic filter to membrane bioreactor. Desalination 276(1–3):272–277. https://doi.org/10.1016/j.desal.2011.03. 062
- Hatree EF (1972) Determination of protein: a modification of the Lowry method that gives a linear photometric response. Anal Biochem 48:422–427. https://doi.org/10.1016/0003-2697(72)90094-2
- Ho JH, Khanal SK, Sung S (2007) Anaerobic membrane bioreactor for treatment of synthetic municipal wastewater at ambient temperature. Water Sci Technol 55(7):79–86. https://doi.org/10.2166/ wst.2007.130
- Hofs B, Ogier J, Vries D, Beerendonk EF, Cornelissen ER (2011) Comparison of ceramic and polymeric membrane permeability and fouling using surface water. Sep Purif Technol 79(3):365–374. https://doi.org/10.1016/j.seppur.2011.03.025
- Huang Z, Ong SL, Ng HY (2011) Submerged anaerobic membrane bioreactor for low-strength wastewater treatment: effect of HRT and SRT on treatment performance and membrane fouling. Water Res 45(2):705–713. https://doi.org/10.1016/j.watres.2010.08.035
- Hubadillah SK, Othman MHD, Tai ZS, Jamalludin MR, Yusuf NK, Ahmad A, Rahman MA, Jaafar J, Kadir SHSA, Harun Z (2020) Novel hydroxyapatite-based bio-ceramic hollow fiber membrane derived from waste cow bone for textile wastewater treatment. Chem Eng J 379:122396. https://doi.org/10.1016/j.cej.2019. 122396
- Hubadillah SK, Jamalludin MR, Othman MHD, Iwamoto Y (2022) Recent progress on low-cost ceramic membrane for water and wastewater treatment. Ceram Int. https://doi.org/10.1016/j.ceram int.2022.05.255
- Jamalludin MR, Harun Z, Othman MHD, Hubadillah SK, Yunos MZ, Ismail AF (2018) Morphology and property study of green ceramic hollow fiber membrane derived from waste sugarcane bagasse ash (WSBA). Ceram Int 44(15):18450–18461. https:// doi.org/10.1016/j.ceramint.2018.07.063
- Jang Y, Kim HS, Ham SY, Park JH, Park HD (2021) Investigation of critical sludge characteristics for membrane fouling in a submerged membrane bioreactor: Role of soluble microbial products and extracted extracellular polymeric substances. Chemosphere 271:129879. https://doi.org/10.1016/j.chemosphere.2021.129879
- Jeong Y, Cho K, Kwon EE, Tsang YF, Rinklebe J, Park C (2017a) Evaluating the feasibility of pyrophyllite-based ceramic membranes for treating domestic wastewater in anaerobic ceramic membrane bioreactors. Chem Eng J 328:567–573. https://doi.org/10.1016/j. cej.2017.07.080
- Jeong Y, Lee S, Hong S, Park C (2017b) Preparation, characterization and application of low-cost pyrophyllite-alumina composite ceramic membranes for treating low-strength domestic wastewater. J Membr Sci 536:108–115. https://doi.org/10.1016/j.memsci. 2017.04.068
- Ji J, Sakuma S, Ni J, Chen Y, Hu Y, Ohtsu A, Chen R, Cheng H, Qin Y, Hojo T, Kubota K (2020) Application of two anaerobic membrane bioreactors with different pore size membranes for



municipal wastewater treatment. Sci Total Environ 745:140903. https://doi.org/10.1016/j.scitotenv.2020.140903

- Kong Z, Li L, Wu J, Wang T, Rong C, Luo Z, Pan Y, Li D, Li Y, Huang Y, Li YY (2021) Evaluation of bio-energy recovery from the anaerobic treatment of municipal wastewater by a pilot-scale submerged anaerobic membrane bioreactor (AnMBR) at ambient temperature. Bioresour Technol 339:125551. https://doi.org/10. 1016/j.biortech.2021.125551
- Lawal NS, Ogedengbe K, Ojo O, Odufowokan AA (2020) Assessment of a submerged membrane bioreactor with composite ceramic filters for cassava wastewater treatment. Res Agric Eng 66(2):72–79. https://doi.org/10.17221/109/2018-RAE
- Li XY, Yang SF (2007) Influence of loosely bound extracellular polymeric substances (EPS) on the flocculation, sedimentation and dewaterability of activated sludge. Water Res 41(5):1022–1030. https://doi.org/10.1016/j.watres.2006.06.037
- Lin H, Chen J, Wang F, Ding L, Hong H (2011) Feasibility evaluation of submerged anaerobic membrane bioreactor for municipal secondary wastewater treatment. Desalination 280(1–3):120–126. https://doi.org/10.1016/j.desal.2011.06.058
- Liu Y, Liu H, Cui L, Zhang K (2012) The ratio of food-to-microorganism (F/M) on membrane fouling of anaerobic membrane bioreactors treating low-strength wastewater. Desalination 297:97–103. https://doi.org/10.1016/j.desal.2012.04.026
- Liu Y, Zhang X, Ngo HH, Guo W, Wen H, Deng L, Li Y, Guo J (2018) Specific approach for membrane fouling control and better treatment performance of an anaerobic submerged membrane bioreactor. Bioresour Technol 268:658–664. https://doi.org/10.1016/j. biortech.2018.08.043
- Liu M, Zhu Z, Zhang Z, Chu Y, Yuan B, Wei Z (2020) Development of highly porous mullite whisker ceramic membranes for oil-in-water separation and resource utilization of coal gangue. Sep Purif Technol 237:116483. https://doi.org/10.1016/j.seppur.2019.116483
- Liu X, Liu J, Deng D, Li R, Guo C, Ma J, Chen M (2021) Investigation of extracellular polymeric substances (EPS) in four types of sludge: factors influencing EPS properties and sludge granulation. J Water Process Eng 40:101924. https://doi.org/10.1016/j.jwpe. 2021.101924
- Mouratib R, Achiou B, El KM, Younssi SA, Tahiri S (2020) Lowcost ceramic membrane made from alumina-and silica-rich water treatment sludge and its application to wastewater filtration. J Eur Ceram Soc 40(15):5942–5950. https://doi.org/10.1016/j.jeurc eramsoc.2020.07.050
- Neto JDSA, de França MJS, de Amorim Junior NS, Ribeiro DV (2021) Effects of adding sugarcane bagasse ash on the properties and durability of concrete. Constr Build Mater 266:120959. https:// doi.org/10.1016/j.conbuildmat.2020.120959
- Nilusha RT, Yu D, Zhang J, Wei Y (2020) Effects of solids retention time on the anaerobic membrane bioreactor with yttria-based ceramic membrane treating domestic wastewater at ambient temperature. Membranes 10(9):196. https://doi.org/10.3390/membranes10090196
- Qin W, Peng C, Wu J (2017) A sacrificial-interlayer technique for single-step coating preparation of highly permeable alumina membrane. Ceram Int 43(1):901–904. https://doi.org/10.1016/j. ceramint.2016.09.206
- Qiu M, Fan Y, Xu N (2010) Preparation of supported zirconia ultrafiltration membranes with the aid of polymeric additives. J Membr Sci 348(1–2):252–259. https://doi.org/10.1016/j.memsci.2009. 11.009
- Rasouli Y, Abbasi M, Hashemifard SA (2019) A new combination of microfiltration, powdered activated carbon and coagulation for

treatment of oily wastewater. Int J Environ Sci Technol 16:5595– 5610. https://doi.org/10.1007/s13762-018-1906-5

- Robles A, Ruano MV, Ribes J, Ferrer J (2013) Performance of industrial scale hollow-fibre membranes in a submerged anaerobic MBR (HF-SAnMBR) system at mesophilic and psychrophilic conditions. Sep Purif Technol 104:290–296. https://doi.org/10. 1016/j.seppur.2012.12.004
- Shafiquzzaman M, Alharbi SK, Haider H, AlSaleem SS, Ghumman AR (2020) Development and evaluation of treatment options for recycling ablution greywater. Int J Environ Sci Technol 17:1225–1238. https://doi.org/10.1007/s13762-019-02537-7
- Shim JK, Yoo IK, Lee YM (2002) Design and operation considerations for wastewater treatment using a flat submerged membrane bioreactor. Process Biochem 38(2):279–285. https://doi.org/10. 1016/S0032-9592(02)00077-8
- Smith AL, Skerlos SJ, Raskin L (2013) Psychrophilic anaerobic membrane bioreactor treatment of domestic wastewater. Water Res 47(4):655–1665. https://doi.org/10.1016/j.watres.2012.12.028
- Tomar SK, Chakraborty S (2018) Effect of air flow rate on development of aerobic granules, biomass activity and nitrification efficiency for treating phenol, thiocyanate and ammonium. J Environ Manag 219:178–188. https://doi.org/10.1016/j.jenvman.2018.04.111
- Umamaheswaran K, Batra VS (2008) Physico-chemical characterisation of Indian biomass ashes. Fuel 87(6):628–638. https://doi.org/ 10.1016/j.fuel.2007.05.045
- Wang Z, Xu Z, Qiu D, Chu Y, Tang Y (2021) Beneficial utilization of Al/Si/O-rich solid wastes for environment-oriented ceramic membranes. J Hazard Mater 401:123427. https://doi.org/10.1016/j. jhazmat.2020.123427
- Wu SC, Lee CM (2011) Correlation between fouling propensity of soluble extracellular polymeric substances and sludge metabolic activity altered by different starvation conditions. Bioresour Technol 102(9):5375–5380. https://doi.org/10.1016/j.biortech.2010. 11.093
- Xiong Y, Harb M, Hong PY (2016) Characterization of biofoulants illustrates different membrane fouling mechanisms for aerobic and anaerobic membrane bioreactors. Sep Purif Technol 157:192–202. https://doi.org/10.1016/j.seppur.2015.11.024
- Yin X, Guan K, Gao P, Peng C, Wu J (2018) A preparation method for the highly permeable ceramic microfiltration membrane–precursor film firing method. RSC Adv 8(6):2906–2914. https://doi.org/ 10.1039/C7RA12314K
- Yue X, Koh YKK, Ng HY (2015) Effects of dissolved organic matters (DOMs) on membrane fouling in anaerobic ceramic membrane bioreactors (AnCMBRs) treating domestic wastewater. Water Res 86:96–107. https://doi.org/10.1016/j.watres.2015.07.038
- Zhang HM, Xiao JN, Cheng YJ, Liu LF, Zhang XW, Yang FL (2006) Comparison between a sequencing batch membrane bioreactor and a conventional membrane bioreactor. Process Biochem 41(1):87–95. https://doi.org/10.1016/j.procbio.2005.03.072
- Zhao C, Xu X, Chen J, Wang G, Yang F (2014) Highly effective antifouling performance of PVDF/graphene oxide composite membrane in membrane bioreactor (MBR) system. Desalination 340:59–66. https://doi.org/10.1016/j.desal.2014.02.022
- Zielińska M, Bułkowska K, Cydzik-Kwiatkowska A, Bernat K, Wojnowska-Baryła I (2016) Removal of bisphenol A (BPA) from biologically treated wastewater by microfiltration and nanofiltration. Int J Environ Sci Technol 13:2239–2248. https://doi.org/10. 1007/s13762-016-1056-6
- Zou D, Qiu M, Chen X, Drioli E, Fan Y (2019) One step co-sintering process for low-cost fly ash based ceramic microfiltration

membrane in oil-in-water emulsion treatment. Sep Purif Technol 210:511–520. https://doi.org/10.1016/j.seppur.2018.08.040

Zuriaga E, Pastor I, Hernández B, Basiero L, Lorente-Ayza MM, Bordes MC, Sanchez E, Abellán M (2017) Low-cost ceramic membranes manufacture for MBR: comparison of pilot and industrial scale. In: Frontiers international conference on wastewater treatment and modelling. Springer, Cham, pp 193–196. https:// doi.org/10.1007/978-3-319-58421-8_30 Springer Nature or its licensor (e.g. a society or other partner) holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.