Different fouling modes of submerged hollow-fiber and flat-sheet membranes induced by high strength wastewater with concurrent biofouling

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Abstract

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Different fouling modes of submerged hollow fiber and flat sheet membranes induced by high strength wastewater with concurrent biofouling

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Abstract

Exploration of two major commercialized flat sheet and hollow fiber membranes in a submerged membrane fungi reactor fed with a synthetic textile wastewater revealed striking difference in extent and mechanism of fouling between the two types, indicating case-specific scope of choice between those for industrial wastewater treatment. The hollow fiber membrane exhibited fouling by cake-layer composed of fungi and starch, intensity being proportional to the operating flux (0.05-0.3 m/d). Conversely, the flat sheet membrane suffered from immediate internal pore-blocking beyond a critical flux of 0.2 m/d. During experiment with major constituents of the synthetic wastewater separately, while media containing only starch and only dye induced negligible fouling, flux-dependent pore-blocking emerged in case of both the hollow fiber (0.288 m/d) and flat sheet membranes (1.3 m/d) for the mixture of starch and dye. Despite a remarkable 99% color and 97% TOC removal achieved by both the membranes, fouling with different mode and intensity for the two types of membranes under similar condition, and for the same type of membrane under different exposure condition warrants development of suitable modules for such recalcitrant wastewater.

Keywords: Submerged membrane bioreactor; Flat sheet; Hollow fiber; Textile wastewater

1. Introduction

Membrane bioreactor (MBR), a remarkable improvement over the conventional activated sludge treatment, offers the advantages of suspended solids and macro-colloidal material free permeate as well as combination of high biomass concentration (requiring small footprint) and complete retention of solids allowing the process to be operated at low organic loading rates, hence, yielding reduced excess sludge production [1]. Submerged membrane bioreactor, since its introduction in 1980s [2] as a cost-effective alternative to sidestream membrane bioreactor (by virtue of eliminating need of recirculation of mixed liquor between bioreactor and membrane unit), over the years has been successfully used to different domestic and industrial wastewater treatment, involving, among others, hollow fiber and flat sheet membranes [3]. Membrane fouling, an inevitable consequence of interactions between membrane and the mixed liquor, however, impedes widespread use of MBR technology. Strategies including development of new membrane materials and membrane modules, modification of feed flow pattern, incorporation of in situ or ex situ cleaning regimes in the membrane unit or a combination therein has been suggested [4] for alleviation of membrane fouling problem.

A submerged membrane fungi reactor, coupling the excellent degradation

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capability (due to non-specific extracellular oxidative enzymes) of white-rot fungi with the inherent advantages of membrane bioreactor, was envisaged as an efficient system for treatment of textile wastewater which is notoriously known as a complex and highly variable mixture of many polluting substances ranging from inorganic compounds and elements to polymers and organic products inducing persistent color coupled with organic load leading to disruption of the total ecological/symbiotic balance of the receiving water stream [5]. The inadequacy, from technical and/or economical points of view, of the conventional biological and physico-chemical treatment systems for textile wastewater, together with the impeded implementation of white-rot fungi in large scale industrial waste treatment due to lack of appropriate reactor system [6] stimulated such a selection. Development of a suitable membrane module, following preliminary trials with the commercially available ones, was deemed to be a crucial initial step towards realization of such a membrane based biological treatment system for textile wastewater. Exploration of two major commercialized flat sheet (Kubota) and hollow fiber (Mitsubishi Rayon) membranes in association with a synthetic textile wastewater revealed striking difference in extent of fouling between the two types, which is in contrary to their usual similar performance under mild operation condition (e.g, sewage treatment) [7].

This paper seeks to elucidate the intriguing disparity of the membrane fouling mechanisms in case of flat sheet and hollow fiber membranes observed under severe operating conditions induced by high strength wastewater with concomitant biofouling. The pollutant removal performance, observed to be influenced to some extent by such distinct fouling mechanisms, have also been accordingly addressed.

2. Materials and methods

This study involved experiments employing a synthetic textile wastewater in submerged microfiltration membrane bioreactor, initially inoculated with pure culture of fungi, but operated, other than controlling pH (4.5±0.2) and temperature (29±1°C), under non-sterile condition. The investigations carried out within the scope of this study may be categorized under two sequential steps. The first step comprises extended period observation, under low operating flux, of the membrane fouling and flux decline along with associated pollutant removal (TOC and color) performance in case of a membrane with horizontally arranged hollow fibers. The subsequent phase, constituting the core of this study, involved further elaborate explorations encompassing mechanism of fouling and effects of varying operating flux and wastewater compositions on fouling along with pollutant removal in case of hollow fiber and flat sheet membranes.

2.1 Microorganisms and chemicals

The white-rot fungi *C. versicolor*, NBRC 9791 obtained from the NITE Biological Resource Center (NBRC), Japan was used for this study. The stock culture was grown, as prescribed by NBRC, on Potato Sucrose Agar (PSA) medium at 26.5°C. The culture was maintained at 4 °C and refreshed every 30–40 days. The decoloration capacity of the fungus strain was confirmed through agar-plate and aqueous batch studies [5].Dye Poly S-119 (polyvininylamine backbone with azo chromophore, orange color) was purchased from Sigma Chemical Co. All other chemicals used were of reagent grade.

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2.2 Synthetic wastewater

The synthetic media used in this study consisted of dye added in the low nitrogen growth media optimized by Kapdan *et al*. [8] for *C.* v*ersicolor* with only modification in that being replacement of glucose by starch, which is used in real textile wet processing as a sizing agent. The media was made of 4.5 gl^{-1}starch, 0.4 gl^{-1} urea, 2 gl^{-1} KH_2PO_4, 0.099 g l^{-1} $CaCl_2$, 1.025 gl^{-1} $MgSO_4.7H_2O$, 0.001 gl^{-1} thiamine, 1 ml l^{-1} trace elements and 100 mg/l dyestuff. The TOC of the medium was around 2000 mg/l (dye TOC≈ 50mg/l). Stock trace elements solution was prepared by dissolving 0.125 g $CuSO_4.5H_2O$, 0.05 g H_2MoO_4, 0.061 g $MnSO_4.5H_2O$, 0.043 g $ZnSO_4.7H_2O$, 0.082 g $Fe_2(SO_4)_3.14H_2O$ in 1 l of milli-Q water.

2.3 Equipment and operating conditions of the bioreactors

2.3.1 Investigation with hollow fiber membrane

A laboratory scale bioreactor, made of PVC, with a working volume of 12.5 liter and containing two hollow fiber polyethylene membranes (UMF 0234L1, Mitsubishi Rayon), each having a surface area and pore size of 0.2 m^2 and 0.4μm, respectively was used for this part of the study. A schematic of the experimental set-up is depicted in Fig. 1.a. Three fine air bubble diffusers - one (air flow 5 l/min), placed in between the two membranes, and the other two, in between each membrane and reactor wall (each 2.5 l/min.), all three supplying air with an intensity of 0.01 m/s, were operated alternately with a 5 min. cycle so that at any time the total aeration rate in the reactor was 5 l/min. Effluent was filtered out through the membranes by suction pumps with a 5 min. on/off cycle (average and instantaneous flux of 0.05 and 0.1 m/d, respectively) for the first 9 days after when the cycle was changed to 9 min. on and 1 min. off to reduce the instantaneous flux (0.055 m/d) while maintaining the same average flux resulting in a HRT of 15 hrs. The transmembrane pressure (TMP) was monitored using vacuum pressure gauges (GC 61, JUST).

2.3.2 Comparison of Hollow fiber and Flat sheet membrane

One hollow fiber (SURM 0334, Mitsubishi Rayon) and one flat sheet membrane (Kubota) both having a pore size of 0.4μm and with respective surface areas of 0.03 m^2 and 0.1 m^2 were submerged in a PVC reactor (working volume=18.75L). Three coarse air bubble diffusers (1 mm perforations), placed as shown in Fig. 1.b, provided continuous aeration with same intensity through the flow channels.

The performances of the two membranes were compared under different operating fluxes (0.05, 0.1, 0.2 and 0.3 m/d) while keeping all other operational conditions (e.g., feed concentration, MLSS concentration of 8 g/L, suction cycle of 9 min. on and 1 min. off, aeration of 0.01m/s) the same. Performances at each flux employing different aeration intensities (0.01-0.03 m/s) were also investigated. After operating the reactor

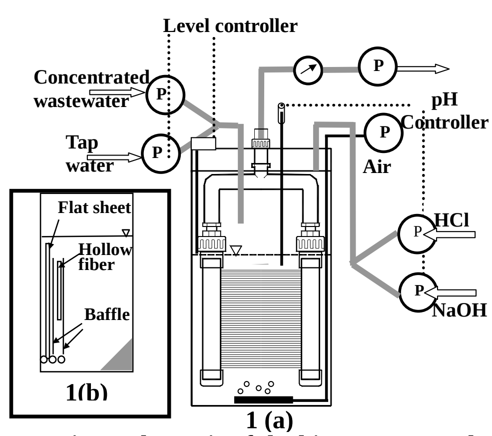

Fig.1 Schematic of the bioreactors used
1(a) Reactor 1
1(b) Side-view of reactor 2

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under a specific flux for a certain period, the membranes were cleaned chemically with NaOCl solution (3% effective Cl content) and reused for experiment under the next elevated flux following confirmation of retrieval of the initial flux.

2.4 Analytical methods

Total organic carbon was measured with a Total organic carbon analyzer (TOC-V, Shimadzu). Color measurements were carried out spectrophotometrically using a spectrophotometer (U-2010, Hitachi) to measure the absorbance of the sample at the peak wavelength of the dye used. The concentration of dyestuff was calculated from a calibration curve of 'absorbance versus concentration' and concentration values were used for calculations of decolorization efficiency. MLSS concentration was measured according to the Standard methods.

3. Results and Discussion

3.1.1 Increase of TMP and flux decline

The TMP rose up to 40 kpa within 5 days, and to 60 kpa within 10 days (Fig.2). The rise in TMP was accompanied with decline in permeate flux (35% in 10 days and 74% in 20 days). Accordingly, higher 'flux per unit pressure' (Fig.2) observed at the initial stage decreased later on and could

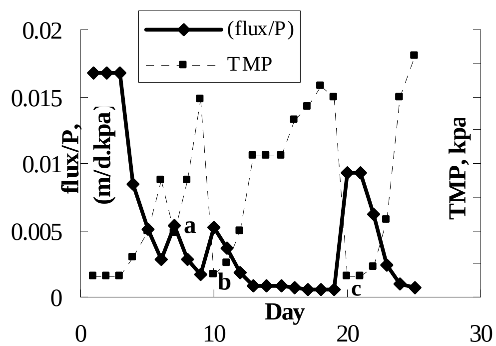

Fig.2 Transmembrane pressure increase and flux decline [a,b,c→ points of flux recovery attempts]

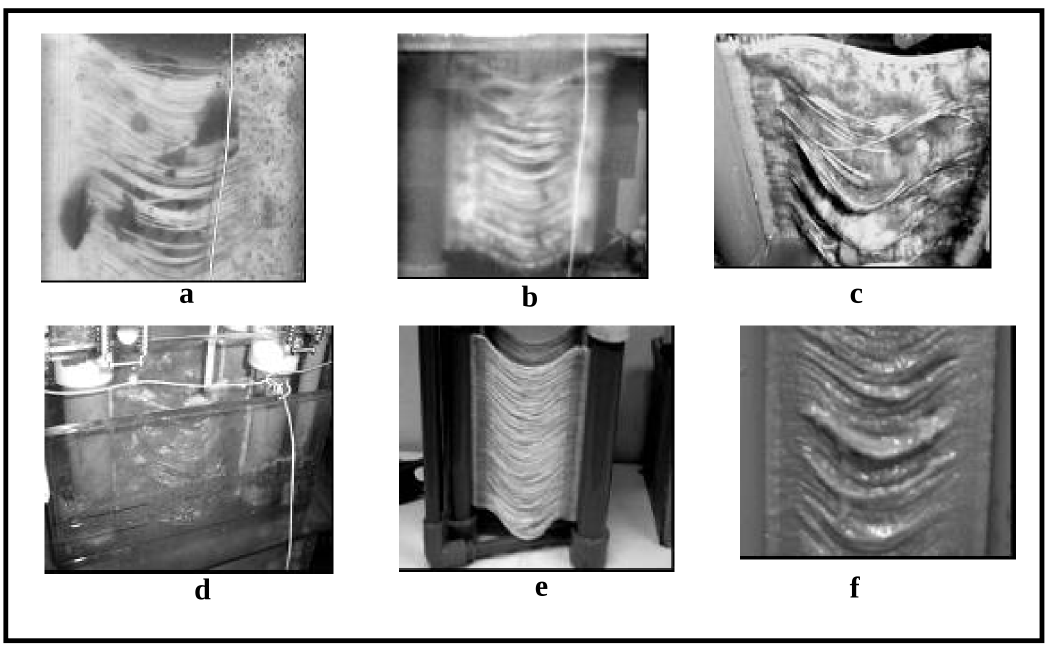

Fig.3 Photographic illustration of membrane fouling

a. Day 1 of acclimation period; **b→f**: During continuous operation (Day 1, 3, 7, 20 & 25); **e**: After cleaning using water (Day 20)

3.1 Investigation with hollow fiber membrane

be recovered only to some extent by *in situ* membrane brushing (day 7, 'a'); suction

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cycle change and sludge withdrawal (day 10, 'b'); and simultaneous *ex situ* cleaning and sludge withdrawal (day 20, 'c').

3.1.2 Mechanism of fouling

The hollow fiber membrane was susceptible to fouling by cake layer composed of fungi and sticky starch. The excessive growth of fungi and, more importantly, their extensive attachment to the membrane fibers on one hand, and the inadequacy of the cleaning strategies adopted, on the other hand, were deemed responsible for the severity of the problem. Figure 3 provides a photographic representation of membrane fouling at different stages of operation period.

In this study, although the initial MLSS concentration of the reactor was less than 2 g/l, it increased gradually and, despite total sludge withdrawal of 7 liters in two steps (on 10th and 20th day), the MLSS concentration rose up to 29g/l within 25 days. In view of the almost similar diminishing rate of flux per unit pressure for the virgin membrane during initial 5 days (low MLSS), and that for the cleaned membrane during the last 5 days (high MLSS), as shown in fig.2, it may be stated that the fungal attachment to the membrane fibers and hence the extent of fouling was independent of MLSS concentration itself, rather more influenced by the inadequacy of the cleaning strategies adopted (e.g., air-scouring, in situ brushing). The filamentous fungi were entangled with the membrane fibers in such a way that the fine air bubbles from the diffusers could not scrub the fungi off the membrane; rather the bubbles sometimes pushed the fungi more into the membrane.

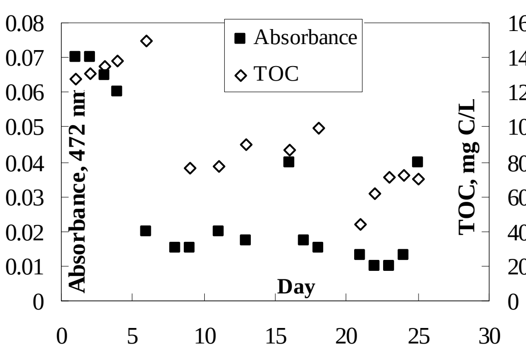

Fig.4 Decoloration and TOC removal performance: First hollow fiber reactor (Initial absorbance=3; TOC= 2g/L)

3.1.3 Pollutant removal

The reactor achieved a 97% TOC removal (initial TOC= 2000 mg C /L), and around 99% color removal (fig.4), with evidence of simultaneous biodegradation of dye (fig.5).

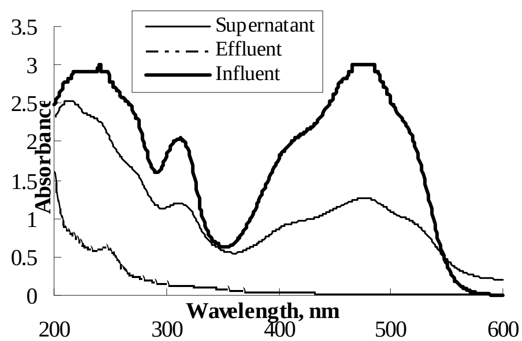

Fig.5 UV-VIS spectra indicating biodegradation of dye

Major portion of the influent TOC was contributed by the high dose of poorly soluble starch, which was observed to be adsorbed on the fungi mass attached on the membrane and, hence, the membrane used in this study (pore size 0.4µm) was able to retain a considerable portion of this starch by sieving. Similarly, synergistic contribution of the microfiltration membrane and fungi-starch cake-layer on it resulted in high retention of the soluble dye, which was subsequently degraded by fungi.

3.2 Comparison of Hollow fiber and Flat sheet membrane

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With the susceptibility of hollow fiber membrane to fouling by cake layer composed of fungi and sticky starch, trial with a different type of membrane was deemed imperative. An experiment was devised to assess the extent of fouling and probable difference in membrane fouling mechanism in case of a hollow fiber and flat sheet membrane under similar operating conditions.

3.2.1 Membrane fouling mechanism and effect of varying flux

The TOC and color removal efficiencies of each type of membrane under different fluxes, and the two types of membranes under similar fluxes were all comparable, and ranged around 97% and 99%, respectively, the quality of the permeate from the hollow fiber membrane being a slightly better as compared to that from the flat sheet membrane (data not shown).

The flat sheet membrane showed stable TOC and color removal under the fluxes 0.05, 0.1 and 0.2 m/d for an observation period of more than 10 days without any increase of transmembrane pressure (fig.6). However, under a flux of 0.3 m/d, immediate severe increase of transmembrane pressure and loss of permeate flux (fig.7) were observed, although the permeate quality was almost the same as that in lower fluxes. The membrane at this point appeared intensely colored; its surface, however, was devoid of any substantial cake-layer (fig.8). Washing with water could neither recover the original appearance of membrane surface or initial flux, nor could it diminish the TMP; consequently, necessitating chemical cleaning (Table.1). Evidently, with the synthetic wastewater used in this study, the flat sheet membrane, beyond a critical flux (0.2 m/d), suffers from immediate internal pore blocking. In analogy to the absence of pore blocking in case of the hollow fiber membrane with a thick cake-layer on it (section 3.1.2), it may be stated that existence of an optimum cake-layer on the flat sheet membrane could be beneficial for avoiding pore blocking. Such layer, as mentioned earlier, could not develop due to smooth membrane surface along with applied strong aeration.

Conversely, in case of the hollow fiber membrane, similar to the observations during the first step of this study, the external fouling due to extensive fungi attachment played the critical role. The transmembrane pressure increased with time (fig.6) and it was accompanied by

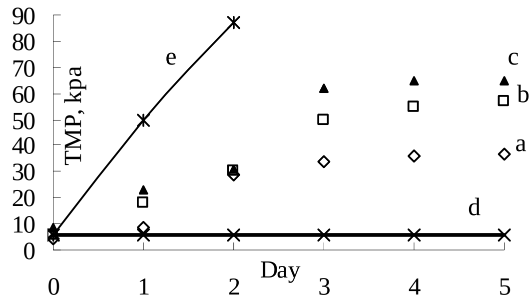

Fig.6 Variation of TMP against Hollow fiber (HF) and Flat sheet (Flat) membrane.
a→c [HF with flux of 0.05, 0.2 & 0.3 m/d];
d→e [Flat sheet with flux of 0.2 and 0.3 m/d]

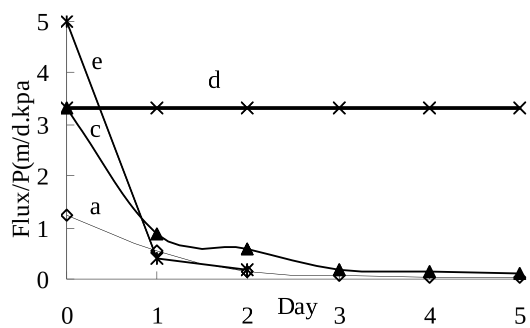

Fig.7 Variation of '[Flux/Pressure]*100' against Hollow fiber and Flat sheet membrane

drop in flux (almost 80% in 5 days, fig.7), the severity increasing with the increase in initial operating flux. It was confirmed that even aeration with a higher intensity (up to 0.03 m/s) could not avoid such fouling. The permeate quality at different fluxes,

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however, was comparable to that in case of flat sheet membrane.

Table 1 TMP values after water and chemical cleaning (Operating flux= 0.3 m/d)

		Initial	Fouling	Cleaning	
				Water	Che-mical
TMP (kpa)	HF	6	65*	10	6
	Flat	7	86#	86	7

*,# corresponding to 5 days & 2 days, respectively (refer to Fig. 6)

3.2.2 Effect of wastewater constituents on fouling

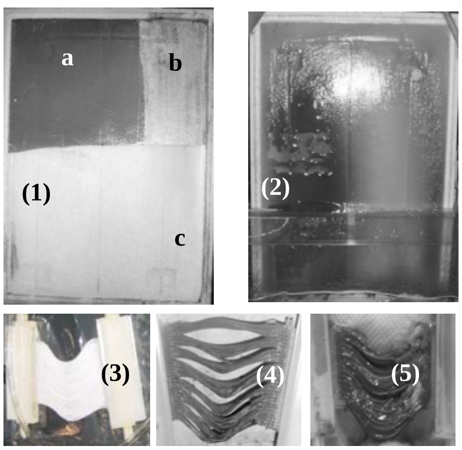

Fig.8 Comparison of fouling
[**Flat sheet membrane** : 1. Pore blocking (dye+ starch mixture) - Fouled membrane surface (a), after tissue -paper wiping (b), fouled spacer visible after removing surface skin (c).
2. Pore blocking in Bioreactor (negligible cake layer)
Hollow fiber : 3.Virgin membrane; 4. Pore-blocking (dye+starch); 5.Cake-layer fouling (Bioreactor)

Since the flat sheet membrane was vulnerable to internal pore blocking, to further elucidate the contribution of the major components of the synthetic wastewater to fouling, the variation of TMP over 24 hrs. (time taken for initiation of severe pore-blocking in case of flat sheet membrane in bioreactor) under a flux of 0.3 m/d was recorded during filtration of media containing only starch, only dye, and mixture of dye and starch,

respectively. Table 2 lists the results from this investigation. The media containing only starch and only dye induced negligible increase in TMP under a flux of 0.3 m/d or many folds of that. For the mixture of starch and dye, however, severe pore blocking (corresponding to a TMP of 50 kpa) emerged in case of both the hollow fiber and flat sheet membranes under respective fluxes of 0.288 and 1.3 m/d (fig.8). This implies a rather different performance of the membranes when exposed to mixed liquor of bioreactor (fungi+wastewater) and to the different constituents of the synthetic wastewater separately. The difference in operating fluxes responsible for initiation of fouling in bioreactor (under 0.3 m/d) and that during filtering mixture of dye and starch (under 1.3 m/d) suggests the combined contribution of starch, dye and fungi particles to pore-blocking of flat sheet membrane in bioreactor. The observed vulnerability of the hollow fiber membrane to pore-blocking during filtering mixture of dye and starch, and not during operation

Table 2. Influence of wastewater constituents on fouling

		Only dye		Starch+ dye		Mixed liquor in bioreactor	
		HF	Flat	HF	Flat	HF	Flat
Fouling	Mechanism	Negligible fouling [a]		Pore blocking		Cake-layer fouling	Pore blocking
	Time #	--	--	24 hr	24 hr	5 days	24 hr
Operating flux *		--	--	0.288 m/d	1.3 m/d	0.1m/d	0.3 m/d
Dye retention		56%	58%	97.7[c]-99[d] %	98 [c] -99 [d] %	99%	99%

[a]up to observation range of 2 m/d; # for TMP rise up to 50 kpa, * causing initiation of severe fouling; [c] before fouling, [d] after fouling

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in bioreactor was simply due to the presence of a cake-layer on the membrane in the later case.

The results from this part of the study when linked with those from the bioreactor experiments, highlights the difference in extent of fouling and associated fouling mechanism for hollow fiber and flat sheet membranes depending on the operational conditions they are exposed to. This, in turn, warrants case specific fouling remediation strategies.

3.2.3 Insight on fouling abatement strategies

Membrane fouling in MBR is an inevitable consequence of interactions between membrane and the mixed liquor. Industrial wastewater, being a complex and highly variable mixture of many polluting substances, may impose aggravated pore-blocking or gelatinous cake-layer fouling depending on the type and arrangement of modules and associated operating parameters. This study casts light on the performance of two commercialized hollow fiber and flat sheet membranes exposed to severe operating conditions induced by textile wastewater. Although the use of the fungal reactor, necessitated by the inadequacy of bacterial degradation of dye, contributed to the uniqueness of the observations made here, these observations may convincingly be generalized as embodiment of cases involving serious bio-fouling. The susceptibility of the hollow fiber membrane to inter-fibral and surface fouling by cake-layer composed of fungi and starch, and that of the flat sheet membrane to pore-blocking by the combined fine particulate foulants (dye+starch+fungi) point to two distinct fouling remediation strategies. Flux-dependent pore-blocking of the flat sheet membrane restricts the application of this membrane to a lower operating flux (0.2 m/d, for wastewater mentioned) which may not satisfy the required treated volume due to the low surface area available for flat sheet modules. Operation under higher operating flux would warrant chemical surface cleaning, frequency of which, in turn, would determine the feasibility of the system. Conversely, to avoid merging of hollow fibers following sludge deposition in between them and simultaneously limit average cake layer thickness on each fiber, a densely packed hollow fiber bundle is recommended. This statement corresponds to the underway investigation in our laboratory, preliminary results from which suggest that, exploitation of only air-scouring may not constitute a sound solution, specially with the horizontally mounted hollow fibers; rather a bundle of fiber, with some additional modifications (improvement in module design), would be suitable for the wastewater mentioned here.

While this study intends to underscore the scope of choice between hollow fiber and flat sheet membranes for industrial wastewater treatment, indiscriminate generalization of the observations is, however, not endorsed. For instance, the performance of hollow fiber membranes with more rugged structure than that of the one used in this study may be different. Also, the pore-blocking observed in case of the flat sheet membrane may be considered as a case-specific problem.

4. Conclusions

In contrary to usual similar performance of commercialized hollow fiber and flat sheet membranes under mild operation condition (e.g, sewage treatment), this paper conveys the observed disparity in extent and

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mechanism of fouling in case of the two types when exposed to recalcitrant industrial wastewater conditions.

The investigations in fungi reactor with synthetic textile wastewater revealed a remarkable 99% color and 97% TOC removal achievement by both the membranes. However, the hollow fiber membrane was susceptible to fouling by cake-layer composed of fungi and starch. Conversely, critical flux (0.3 m/d)-dependent pore-blocking of the flat sheet membrane was detected. When exposed to the major constituents of the synthetic wastewater separately, rather different performances of the membranes were observed. While the media containing only starch and only dye induced negligible increase in TMP, flux-dependent pore-blocking emerged in case of both the hollow fiber (0.288 m/d) and flat sheet membranes (1.3 m/d) for the mixture of starch and dye. However, the difference in operating fluxes responsible for initiation of fouling in bioreactor (0.3 m/d) and that during filtering mixture of dye and starch (1.3 m/d) suggested the combined contribution of starch, dye and fungi particles to pore-blocking of flat sheet membrane in bioreactor.

The performance evaluation of commercialized hollow fiber and flat sheet membranes under recalcitrant industrial wastewater condition revealed the necessity of distinct fouling remediation strategies as well as the importance of development of new membrane modules for such wastewater.

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