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Municipal wastewater reclamation for non-potable use using hollow- fiber membranes

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Abstract. Approximately 80% of water used in urban areas reappears as municipal wastewater (MWW). Reclamation of MWW is an attractive proposition under the present scenario of water stressed cities in India. In this paper, we attempted to reclaim MWW using lab-scale hollow- fiber (HF) membrane modules for possible reuse in non-potable applications. Experiments were conducted to evaluate the efficiency of virgin HF (M_1) and modified HF (M_2) modules. The M_2 module consists of HF modified with a skin layer formed through interfacial polymerization of m-phenylenediamine with trimesoyl chloride (MPD-TMC). The molecular weight cut-off (MWCO) of M_1 was 44000 g/mol and that of M_2 10000 -14000 g/mol on the basis of rejection of polyethylene glycol. The combination of M_1 and M_2 modules was able to reduce concentrations of most of the pollutants in sewage and improved the treated water quality to the acceptable limits for non potable reuse applications. It is found that about 98-99% of the initial flux is recovered by the backwashing process, which was approximately two times in a month when operated continuously.

Keywords: sewage reclamation; hollow fiber; ultrafiltration; non potable water; reuse.

1. Introduction

The total volume of the water on the earth is approximately 1.3×10^9 km^3, but only 4 million km^3, *e.g.*, 0.3% of the total water volume is actually usable by mankind (Koltuniewicz and Drioli 2008). In the recent past, human consumption of water has increased dramatically worldwide. It was observed that at least 40% of the world's population will live in the countries that suffer from a chronic shortage of water. Today, already more than 50 countries suffer from a shortage of water but by 2025, two-third of the world population will not have enough drinking water, as per the prediction of the United Nations. Therefore, it is necessary to conserve, recycle and reuse the limited water resources and to develop advanced technologies for treating and reclaiming wastewater from different sources.

The most important role in water recovery, reuse and recycling is to control composition of water streams. There are two main options to this end i.e. conservative separation for recovery (*e.g.*, filtration) and/or destructive conversion (*e.g.*, advanced oxidation) for removal of various substances from

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water. Membranes play an important role in both of these methods and therefore attracted attention for reclamation of municipal wastewater as it is highly efficient, economical and easy to operate (Lens *et al*. 2002, Quin *et al*. 2004, Bixio *et al*. 2005, Wntgen *et al*. 2005). Generally, a dual membrane processes such as MF or UF and RO is becoming increasingly attractive as a technology for the reclamation of municipal wastewater (Tam *et al*. 2007).

In this paper, we attempted the use of HF membrane modules for reclamation of municipal wastewater. Two HF modules, one of which contained HF modified with metaphenylene diamine and trimesoyl chloride (MPD-TMC) polymer, were used separately and in tandem to obtain reclaimed water from MWW. The thin film composite HF having polyamide skin layer on the surface of the polysulphone fiber were made by interfacial polymerization of trimesoyl chloride in hexane and m-phenylenediamine in water. The objective of this work was to explore the use of virgin HF and modified HF in that order to reduce the organic matter, nutrients and microorganisms from MWW to obtain reclaimed water for reuse purpose.

2. Material and methods

2.1 HF (Msubscript1) and modified HF membrane (Msubscript2) modules

The hollow fiber membrane modules used in the present study were supplied by CSMCRI, Bhavnagar under CSIR Network Program (NWP-47). The details of preparation of UF-HF fibers and skin layer coating were presented elsewhere (Cadotte 1981, Verissimo *et al*. 2005, Ray *et al*. 2010). The skin layer coating comprises the following steps: passing the fiber in a continuous running mode through the aqueous meta-phenylene diamine solution (0.05/0.1% w/v) solution for about 30 sec, and then through trimesoyl chloride solution (0.01-0.1% w/v in hexane) for 30 sec, and then curing the fibers at 40^{o}C. This results in the formation of stable polyamide layer on the fiber surface. Different hollow fiber membrane modules varying in their length from 32-33 cm and diameter from 1.6 cm to 3.8 cm containing fibers from 100 to 400 were made using commercially available epoxy resin as the potting material.

The details of the two different modules are presented in Table 1. Both modules contain polysulfone based HF, but the modified HF module had fibers coated with an over layer of polymer made of

Table 1 Specifications of HF (M_1) and MHF (M_2) membrane modules

Sr. No.	Specifications	M_1	M_2
1	Length (cm)	32.5	33
2	Diameter (cm)	3.8	1.6
3	No. of fibers	400	100
4	Exposed fiber length in module (cm)	0.25	0.295
5	Total surface area (m^2)	0.3140	0.09263
6	Material	PS	MPD-TMC / PS
7	MWCO (g mol^{-1})	44000-94000	10000-14000
8	Calculated* pore size (nm)	5-7	2-3

*R (pore radius, nm) = $0.045\times(\text{MWCO})^{0.44}$

metaphenylene diamine (MPD) and trimesoyl chloride (TMC). These modules are referred as M_1 and M_2, respectively in the text. This layer effectively contributes to reduction in pore size comparable to that commonly found in the flat sheet RO membranes (Cheryan 1998, Shah and Joshi 2009). In the present study, M_2 contained HF membrane that has 4× lower MWCO and smaller pore size (2-3 nm) relative to those in M_1.

2.2 Experimental setup and operating conditions

The MWW sample was collected from a sewage channel passing through NEERI campus. Fresh sample was collected on each day for experimental studies. The treatment scheme of filtration process is shown in Fig. 1. The MWW sample was filtered through muslin cloth (pore size = 0.1 mm) to remove larger particulate and slimy flocs, prior to application using M_1 and M_2. The HF membrane module was operated in out-to-in mode at 10 psi under cross-flow conditions. The feed was transferred to module through RO booster pump (Model 1800, KEMFLOW, rated flow, 1.8 L min^{-1}). The permeate of HF module (M_1) was considered as feed for modified HF module (M_2). The operating pressure and feed rate was kept constant *i.e.*, 10 psi and 1.8 L min^{-1}, respectively. A two – min. backwashing was given to M_1 and M_2 modules, by reversing the mode of operation *i.e.*, in-to-out mode. The modules were subjected to backwashing with 0.2% citric acid (1×) followed corresponding permeate (2×) and finally Millipore water (2×). The sequence of backwashing was adequate to recover the flux through the modules.

2.3 Analytical methods

Chemical oxygen demand (COD), total dissolved solids (TDS), total suspended solids (TSS), chloride and hardness were determined as per the Standard Methods [APHA 2005]. The total bacterial count was measured in terms of cfu/ml by Pour - Plate method (Method No.:9215B, Standard Methods) and turbidity was measured by using ELICO Nephelometer (CL 52D, Hyderabad). Pore morphology of the membranes was studied using LEO 1400 SEM microscope at 5-15 kV accelerating voltage. MWCO values were measured by passing aqueous solutions of polyethylene glycol (PEG) (M_w 3, 10, 20 and 35 KDa), dextran (M_w 70 KDa) and Polyethylene oxide (M_w 100KDa) each of 500 ppm through the virgin hollow fiber modules at 30 psi by out-to-in flow mode. The percent organic solute in feed and permeate were analyzed by GPC (Waters) using ultrahydrogel columns.

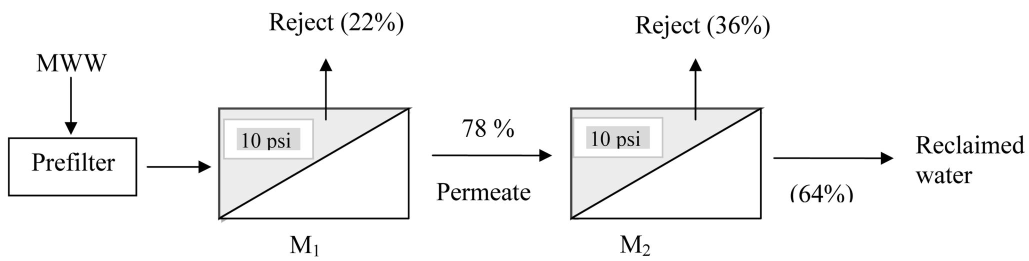

Fig. 1 Flow chart for reclamation of MWW using hollow fiber membrane modules

3. Results and discussion

SEM photomicrographs clearly show that the fibers have asymmetric structure with a dense skin layer on both the inner as well as the outer surface with finger like channels in the middle (Fig. 2a). The finger like channels from inner and outer layers is separated by a sponge like intermediate structure. It is expected that the sponge like intermediate structure provides greater strength for the fiber and hence the durability for long term applications. Fig. 2(b) shows skin layer thickness to be approximately 2 μm and a gradual enhancement in pore size from skin to the interior part of the membrane.

MWCO (molecular weight cut-off) of the virgin membrane was determined using rejection factor (R_f) of polymer solutes like PEG, Dextran and PEO, determined from the experiments of size exclusion chromatography technique. MWCO estimated based on only PEGs is 44000 g/mol while it is 94000 g/mol if considered a combination of PEG, Dextran and PEO solutes (Ray *et al*. 2010). The membrane pore radii calculated are about 5-7 nm. Similarly, MWCO of the modified membranes were determined using rejection factor (R_f) of PEG solutes, determined from the experiments of size exclusion chromatography technique. MWCO estimated based on the PEGs is about 10000-14000 g/mol for the modified membranes which is about one-fourth of the MWCO of virgin membrane.

The M_1 and M_2 modules were tested initially for assessing the hydraulic characteristics (Table 2). In general, water flux is higher than MWW flux for both the modules implying the constituents of MWW can influence practical flux rates. The water flux of M_1 was 5 times higher than that of M_2, whereas it was 7-8 times in case of MWW. The difference could be attributed to different membrane

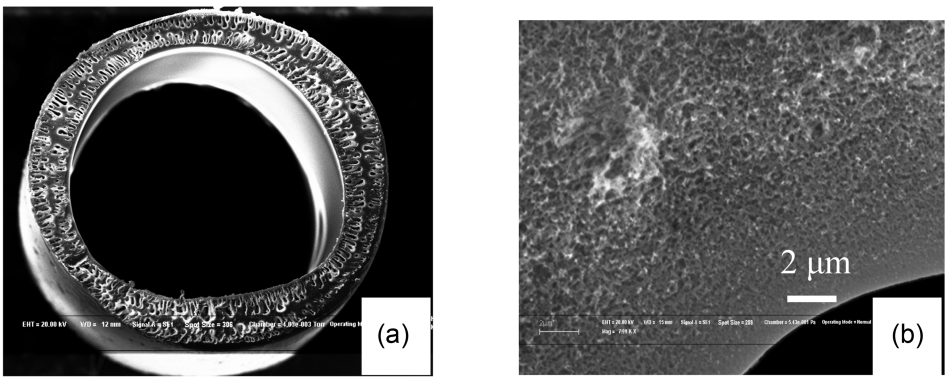

Fig. 2 SEM micrograph of the membrane showing circular bore (a) and (b) gradual variation in pore size from skin to the interior part of the membrane obtained using 20% Polysulphone+2% PVP solution, DMF +water (10/90 v/v) as pore former

Table 2 Cross flow velocity, pure water flux and sewage permeate flux for M_1 and M_2 at 10 psi, and feed flow rate 1.8 L min^{-1}

Membrane module	CFV* ($\times 10^{-5}$ m/s)	Pure Water Flux ($Lm^{-2}h^{-1}$)	Sewage Permeate Flux ($Lm^{-2}h^{-1}$)
M_1	9.55	187.26	154.77
M_2	32.38	37.83	19.91

*CFV = Cross flow velocity (m/s) calculated from: $(L/1000)\times(m^{-2})\times(h^{-1}/3600)$

Table 3 Permeate quality using Msubscript1 and Msubscript1+Msubscript2 systems and Standards for potable water

Sr. No.	Parameter*	Feed quality** (sewage)	Permeate quality		Standards for potable water (IS:10500)
			M_1	M_1+M_2	
1	pH	7.4-7.6	7.5	7.4	6.5-8.5
2	TSS	59-70	16	BDL	----
3	Total Solids (TS)	388-560	341	270-289	500
4	Turbidity	89.9-110	16.5	5.0-9.8	5.0
5	COD	172-236	209	30-45	----
6	Total ammonia	25.5-30	11.9	2.0-5.6	----
7	Total hardness	190-210	190	160-170	300
8	Chloride	160-250	55	40-50	250
9	Total bacterial count (cfu/ml)	75000-2,00,000	23,000	90-110	0.1-0.3

*All values expressed in mg/l except pH and Total bacterial count, **Before pretreatment with muslin cloth
BDL = Below Detectable Limit

resistance of M_1 and M_2. The HF fibers in M_2 carry thin film composite layer as described previously which offers more resistance.

3.1 HF membrane filtration of MWW - permeate quality

Table 3 presents concentrations of various water quality parameters of raw feed (MWW), and permeate of M_1, M_1+M_2. The permeate quality is compared with IS: 10500 Standards for potable water. The raw feed (prior to filtration through muslin cloth) contained the suspended solids, colloidal matter and very large amount of microorganisms. The pre-filtration through muslin cloth did not alter the feed quality significantly except that TSS was slightly less, in the range 50-55 mg/l. The M_1 module removed a part of the suspended solids, colloidal materials and microorganisms. Both TSS and TS were reduced to 16 and 341 mg/l, respectively. The chloride and turbidity was also reduced to 55 mg/l and 16.5 NTU, respectively. The total bacterial count was reduced to 23,000 cfu/ml from 2,00,000 cfu/ml. The results indicate that permeate of M_1 falls short of Standards for potable water with respect to turbidity, TSS, COD, ammonia, and bacterial counts. The performance of M_1+M_2 was evaluated as per the flow chart illustrated in Fig. 1 (Table 3). The permeate quality was good and the levels of most of the pollutants were very low. The operating pressure and permeate flux was shown in Table 2. The concentrations of COD and NH_4^+-N were considerably reduced when both M_1 and M_2 are operated in tandem. The chloride concentration was reduced to <50 mg/l. The permeate from tandem operation (M_1+M_2) showed very remarkable reduction in total bacterial count (≤90-110 cfu/ml). Fig. 3 displays permeate quality in terms of variation in concentrations of observed parameters studied over one-month period. The raw effluent characteristics TS, TSS, TH, Cl- fluctuated within 10% of the average values (Table 3), while total bacterial count on some days during study period showed even greater changes (up to 50% of average value). The data in Fig. 3 indicate that despite fluctuations in the feed water quality, the permeate quality is reasonably stable during the study period. This confirms that the dual membrane process (M_1+M_2) filtration assembly performs consistently and the process can accommodate reasonable variations in the concentrations of the target parameters of the feed. The profile of flux as a function of time for both

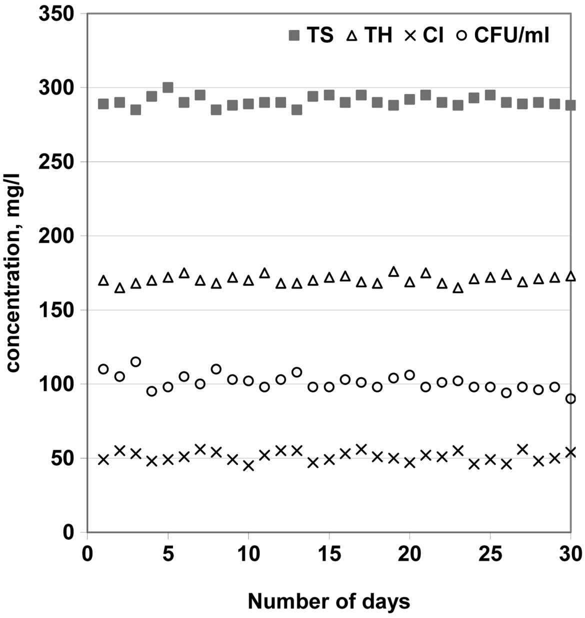

Fig. 3 Permeate quality in terms of concentrations of observed parameters over one month indicating stable performance of the M_1+M_2 filtration assembly

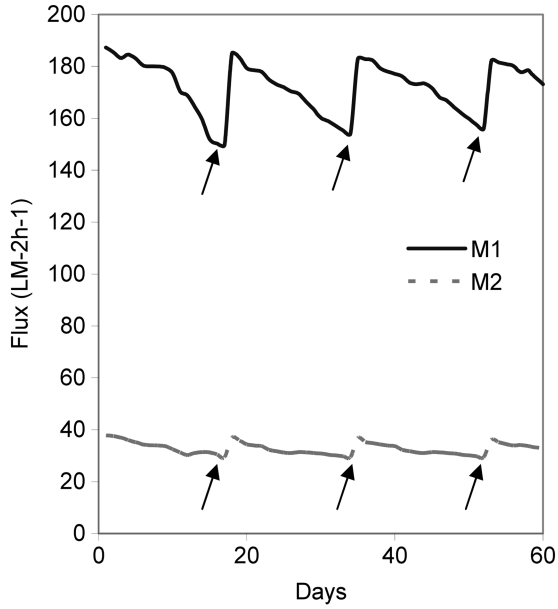

Fig. 4 Time course variation in flux and its regaining through backwashing sequence (arrows denote the day on which backwashing was given)

the modules is shown in Fig. 4. When the permeate flux was reduced to about 80% relative to the initial flux, both modules were backwashed for 2 min by reversing the mode of operation, *i.e.*, in-to-out mode as described in the experimental section. The time course variation of flux for both the modules along with flux recovery upon backwashing is depicted in Fig. 4. It is found that about 98-99% of the initial flux is recovered by the backwashing process. In the present system, the need for backwashing was low, approximately two times in a month when operated continuously. The permeate quality in terms of turbidity, organic content, ammonia, hardness, TSS, TS and total bacterial count could meet the water quality requirements for many reclaimed and non-potable reuse applications. On the basis of results obtained, it is proposed that HF modules can be used for reclamation of sewage. The HF modules required 10 Psi for achieving significant permeate flux (cf. Table 2). The required pressure is two orders of magnitude low compared to the conventional RO membranes which require up to 500-1000 Psi. Due to residual COD, ammonia, and bacterial counts in excess of Standard values for potable water (Table 3), it may be said that the present system does not produce potable quality water, however, the permeate fits to non-potable quality water for several reuse applications.

The reclamation of sewage effluent as a secondary source of water for non-domestic use is a national strategy of Singapore as water is a scarce resource in Singapore. A demonstration plant of a dual MF/RO membrane process with the capacity of 10,000 m^3/day for production of potable grade water from the secondary treated domestic sewage effluent has been successfully operated (Tan and Lee 2001). Koning *et al*. (2008) assessed various water treatment technologies for reuse including those of membrane filtration processes. Childress *et al*. (2005) discussed correlation between HF membrane and module properties, and attributed performance factors to the physical integrity of the membranes. In this study, the combination of HF with modified HF used for reclamation of sewage (Fig. 1). The suspended solids, colloidal matter and some microorganisms are removed by $M_{1,}$ while M_2 removed the organic compounds and most of the residual microorganisms from sewage. Here M_1 served as the pretreatment for modified HF (M_2) to reduce the fouling rate. The hollow fiber configuration have three major advantages (Qin *et al*. 2003): (1) HF modules have much larger ratio of membrane area to unit volume compared to flat and spiral-wound modules and hence higher productivity per unit volume of membrane module; (2) they are self supporting which can be back washed to recover the flux; and (3) they have good flexibility in the mode of operation. However, the present system comprising virgin and modified HF membranes requires more investigations with larger module sizes for longer period before their practical utility can be established. Due to lower pressure application required for the HF modules, the treatment process explored in the present study may be cost effective.

4. Conclusions

On the basis of the experiments with the lab-scale HF modules, the use of hollow fiber for reclamation of sewage can be regarded as technically feasible. The filtration of sewage using HF and modified HF modules in series can produce treated water whose quality permits its reuse in several non-potable applications.

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