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Characteristics of foulants of air-sparged side-stream and sub-merged membrane used to treat municipal wastewater

Asiful Hoque

Department of Civil Engineering
Chittagong University of Engineering & Technology, Chittagong 4349, Bangladesh

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Abstract

Membrane bioreactors (MBRs) have become very attractive during the past decade due to their advantages, although optimization of MBR operation has not been achieved yet. Recently, air-sparged side-stream MBRs (ASMBRs) has received much attention since they can overcome the drawbacks of submerged MBRs such as difficulty in cleaning membrane modules. Widespread application of MBRs has been limited by problems associated with membrane fouling and ASMBRs are not exceptions. A comparison between side-stream and sub-merged MBRs in terms of fouling was carried out in this study. For this purpose, tiny-scale hollow fiber and flat sheet membranes in sub-merged configuration along with tubular membrane in side-stream configuration were examined using same mixed liquor suspension in this study. Membrane flux was set at 42 L/m2/hour for the three types of membranes. Foulants were extracted from the membranes after 40 and 20 days of continuous operations in two experiments. The flat sheet (MF) membrane fouled more rapidly compared to other two types membranes. Tubular membrane was fouled the least among the three types of membranes. Humic like substances were found to be dominant in tubular and flat sheet membranes, whereas, protein was found to be dominant in the hollow fiber membrane.

Keywords: Side-stream, sub-merged, fouling, humic substance, protein.

1. Introduction

Membrane bioreactors (MBRs) are widely applied for wastewater treatment for its several advantages, such as high biodegradation efficiency and high pollutant removal (Le-Clech et al. 2003; Kimura et al. 2004, 2005, 2008). In the history of MBR, different side-stream MBRs have been developed that can be operated with high flux. The main objective was to reduce fouling through high liquid velocities inside tubular membranes (up to 5 m/s). This design has been criticized later because of its high energy consumption. However, such systems are still widely operated in the field of industrial effluent treatment (Le-Clech et al. 2006). To overcome the high energy consumption of side-stream MBRs, submerged MBR were developed which can be operated with lower flux compare to side-stream MBR system. Recently, a third generation MBR, i.e., air-sparged side-stream MBR (ASMBR) was introduced in the field of MBRs. The basic flow pattern of ASMBR is the same as that of first generation side-stream MBRs: vertical tubular membranes are installed outside the bio-reactor and mixed liquor suspension is re-circulated in the membrane modules. Generally, tubular membranes are used in side-stream MBRs whereas hollow fiber and flat sheet membranes are used for submerged type configuration.

Despite a large number of studies on MBR systems, membrane fouling remains the major issue and is still need to be investigated intensively. The influence of operating conditions on sludge properties and, consequently, on fouling has already been reported, mainly for submerged MBRs (Lee et al. 2003; Le-Clech et al. 2006). Still, there are few studies that carry out both for side-stream and submerged MBRs fouling by using real w both side-stream and submerged MBRs. To compare the characteristics of foulant of air-sparged side-stream MBR and submerged MBR for treating the same real municipal wastewater will be interesting.

2. Materials and methods

2.1 Pilot-scale ASMBR

Figure 1 shows a schematic diagram of the experimental unit used in this study. The bioreactor was divided into two zones by inserting baffles so that the system was used as a baffled membrane bioreactor (BMBR). Effective volume of the reactor was 1000 L and the volume ratio of the outer zone/inner zone was 1.5. Two separate tubular membrane modules were connected to the bioreactor and mixed liquor suspension was circulated between the reactor and the modules (Figure 1). The material of the membrane used in this study was polyvinylidene fluoride (PVDF) and the nominal pore size was 0.03 µm for tubular membrane. Each membrane module was comprised of about 100 vertical tubes, having a membrane area of 1.6 m^2. The length and internal diameter of each tube were 1 m and 5.2 mm, respectively. Membrane filtration was carried out in the inside-out mode, and permeate was therefore collected from the outside of the membrane module by a suction pump. To compare the fouling in both side-stream and sub-merged MBR configurations, tiny-scale hollow fiber membrane module (membrane area: 0.011 m^2) and flat sheet membrane module (0.02 m^2) were submerged in the inner zone of the biological reaction tank of the ASMBR. The material of both hollow fiber membrane and flat sheet membrane were PVDF.

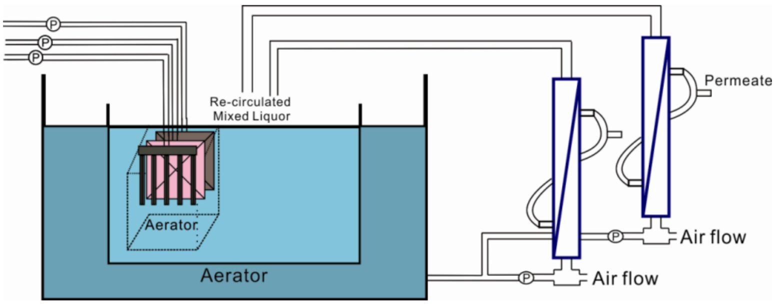

Fig. 1. Schematic diagram of experimental set-up.

2.2 Operational conditions

Two experiments (designated Run 1 and Run 2) were carried out in this study. Run 1 and 2, were carried out for 40 days and 20 days, respectively. In the continuous operation, fifteen-second backwash was carried out every ten minutes of filtration at 100 kPa with permeate.

The high membrane flux could not be applied to submerged membranes as membrane fouling occurred very rapidly. Thus, membrane flux in Run 1 and 2 were set at 42 LMH for all types of membranes. Intermittent operation (1-minute pause in every 10-minute filtration) was carried out for sub-merged MBRs. Operational conditions in Runs 1 and 2 are summarized in Table 1.

Table 1
Experimental Conditions

		Run 1			Run 2			
Membrane flux	($m^3/m^2/day$) (LMH)	1.0 42	1.0 42	1.0 42	1.0 42	1.0 42	1.0 42	1.0 42
Membrane configuration		Side-stream	Submerged	Sub-merged	Side-stream	Sub-merged	Sub-merged	Sub-merged
Membrane Type		Tubular PVDF	Flat Sheet PVDF	Hollow Fiber PVDF	Tubular PVDF	Flat Sheet PVDF	Flat Sheet PVDF	Hollow Fiber PVDF
Nominal pore size	(μm)	0.03	0.1 (MF)	0.4	0.03	0.01 (UF)	0.1 (MF)	0.4
Membrane area	(m^2)	1.6	0.02	0.011	1.6	0.02	0.02	0.011
Filtration		10 min Filtration/ 1 min backwash	10 min Filtration/ 1 min pause	10 min Filtration/ 1 min pause	10 min Filtration/1 min backwash	10 min Filtration/ 1 min pause	10 min Filtration/ 1 min pause	10 min Filtration/ 1 min pause
Experimental Duration	(Days)	40	40	40	20	20	20	20
HRT	(hour)	4.25	4.25	4.25	4.25	4.25	4.25	4.25
SRT	(Days)	40	40	40	40	40	40	40

2.3 Extraction of foulants from the fouled membranes

At the end of the operation period of run 1 and 2, foulants were desorbed from the fouled membranes and analysed. The surface of the membrane surface was manually wiped with a sponge to eliminate the influence of residual deposits in the extraction process. Amounts of the deposits were minimal on the basis of visual inspections. Foulants were extracted by soaking the fouled membranes in a sodium hydroxide solution (pH=12) for 24 hours at 30 ˚C.

2.4 Analytical methods

Total organic carbon (TOC) concentration was determined using a TOC analyser (TOC-VCSH, Shimadzu, Kyoto, Japan). The phenol–sulfuric acid method (Dubois et al. 1956) and the Lowry method (Lowry et al. 1951) were used for determining the concentration of polysaccharide and proteins, respectively. Glucose and bovine serum albumin (BSA) were used as standards for the measurements of polysaccharide and proteins respectively. For FTIR analyses, KBr pellets containing 0.25% of the sample were prepared and examined in an FTIR spectrophotometer (FTIR-8400S Shimadzu Kyoto Japan) at a resolution of 4 cm−1. Fourier transform infrared (FTIR) spectra of the surfaces of physically cleaned membranes were obtained by using an attenuated total reflection (ATR)-FTIR spectrophotometer (FTIR-8400S Shimadzu Kyoto Japan). A fluorescence spectrophotometer equipped with a 150-W ozone-free xenon lamp (RF-5300PC Shimadzu Kyoto Japan) was used for measuring EEM spectra. In fluorescence measurements, the wavelength of both emission and excitation was varied stepwise by 5 nm.

3. Results and discussion

3.1 Changes in Filtration Resistance Observed in the Continuous Operations

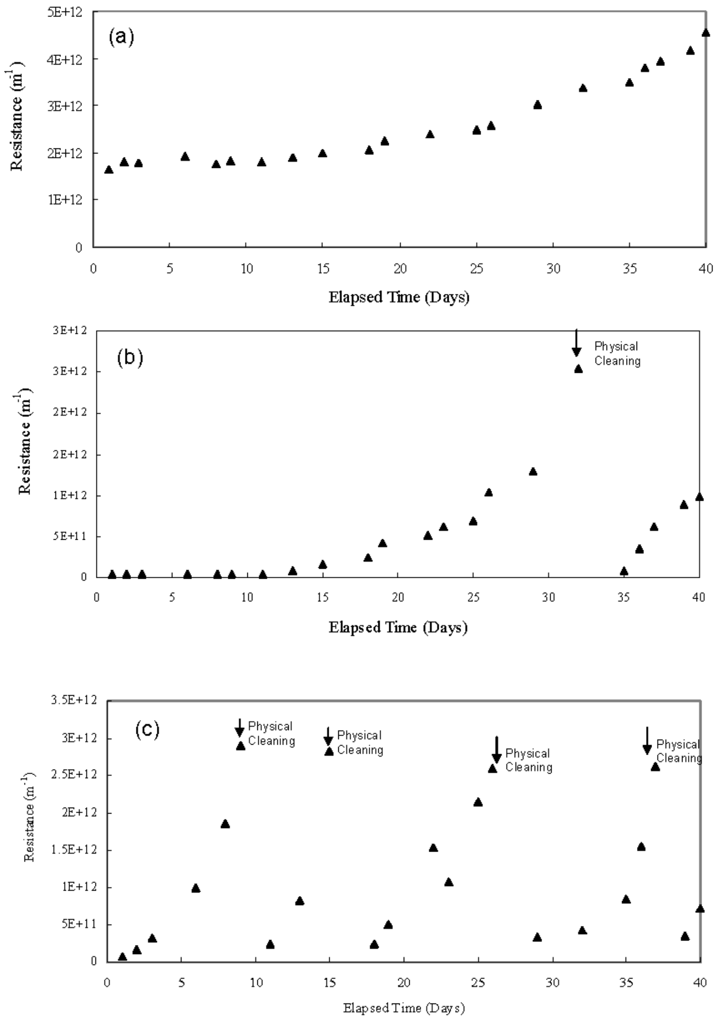

Fig. 2. (a) Change of filtration resistance during Run 1 for tubular membrane, (b) hollow fiber membrane, (c) flat sheet membrane.

Figure 2 shows time course change of resistance in different types of membrane observed in Run 1. In the case of tubular membrane initial resistance of the membrane was higher compare to hollow fiber and flat sheet membranes. In the case of side-stream tubular membrane mixed liquor is re-circulated to the membrane module and inlet pressure of the module cause the higher initial resistance.

However, resistance did not increase significantly in the first 30 days of operation which shows the suitability of ASMBRs for longer period of operation. Figure 2(b) shows time course change of resistance of hollow fiber membrane in Run 1. Initial 10 days of operation, the membrane was not fouled. Therefore, no significant increase in resistance is shown. Resistance started to increase after 10 days of operation and physical cleaning carried out at around 30 day due to flux decline. Time course of resistance development of flat sheet membrane was shown in Figure 2(c). Flat sheet membrane fouled very rapidly and need to clean physically very frequently. Almost every 7 days need to clean flat sheet membrane physically by sponge.

Time course change of resistance in Run 2 was shown in Figure 3. In Run 2, tubular, hollow fiber (MF), flat sheet (MF) and flat sheet (UF) membranes were operated for 20 days using same mixed liquor suspension for direct comparison among these four types of membrane. Figure 3(a) shows the time course change of resistance of tubular membrane in Run 2. The trends of the time course change of resistance of tubular membrane in both run almost having little high initial resistance and not rapid increase of resistance. Figure 3(b) shows the change of resistance for hollow fiber membrane in Run 2.

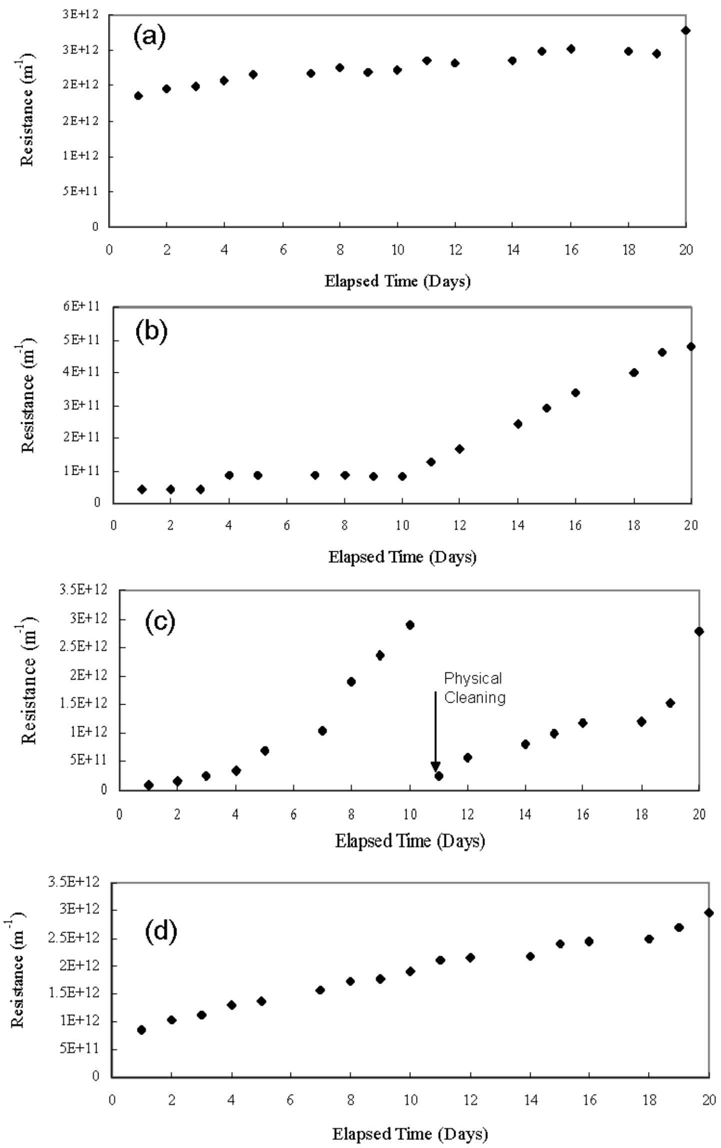

Fig. 3. (a) Change of filtration resistance during Run 2 for tubular membrane, (b) hollow fiber membrane, (c) flat sheet membrane (MF), (d) flat sheet membrane (UF).

First 10 days of operation, resistance was almost same and after that resistance starts to increase. Operation of hollow fiber membrane was carried out for 20 days without any physical cleaning. Figure 3(c), (d) show the time course change of resistance of flat sheet (micro-filtration, MF) and flat sheet (ultra-filtration, UF) membranes, respectively. Flat sheet (MF) membrane fouled very rapidly same as Run 1, whereas Flat sheet (UF) membrane did not fouled rapidly although initial resistance was very high. We used flat sheet (MF) and flat sheet (UF) membranes from two different companies. Flat sheet (MF) membrane used in the experiment was commercially available membrane and flat sheet (UF) membrane was not available commercially. Although both are PVDF membranes but other properties might be different.

3.2 Analysis of foulants

Figure 4 (a) shows the amounts of foulants extracted after Run 1. Amount of organic matters extracted from flat sheet membrane in Run 1 was high, which implies severe fouling of flat sheet membrane. Airlift membrane has the least foulant desorbed from the membranes among the three types of membranes. Figure 4 (b) shows the amounts foulants extracted membrane after continuous operation in Run 2. In Run 2, Flat sheet (MF) membrane also produced more organic foulants compare to other three types of membrane. The extracted organic foulant from flat sheet (UF) membrane was not high as Flat sheet (MF) membrane.

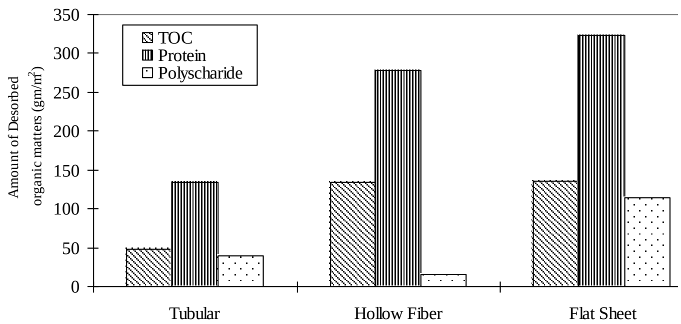

Fig. 4. (a) Amount of desorbed organic matter after Run 1.

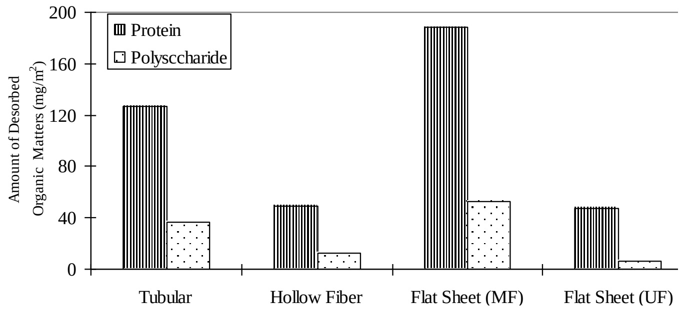

Fig. 4. (b) Amount of desorbed organic matter after Run 2.

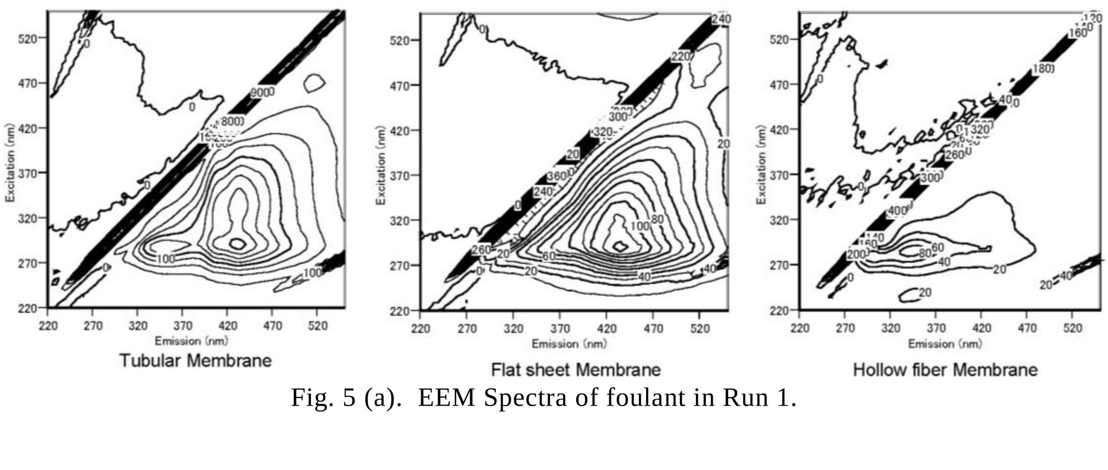

Fig. 5 (a). EEM Spectra of foulant in Run 1.

Figure 5 (a) shows EEM fluorescence spectra measured for the foulants extracted from the fouled membranes at the termination of Run 1. The EEM method is useful for distinguishing different types of organic matter. For tubular and flat sheet membrane, distinguished peaks at 280 nm/430 nm (Ex/Em) and was found in the EEM measured for the extracted foulants and can be attributed to humic substances (Chen et al. 2003). In the EEM of the foulant of hollow fiber membrane shows a peak at 280 nm/350 nm (Ex/Em) which is protein peak.

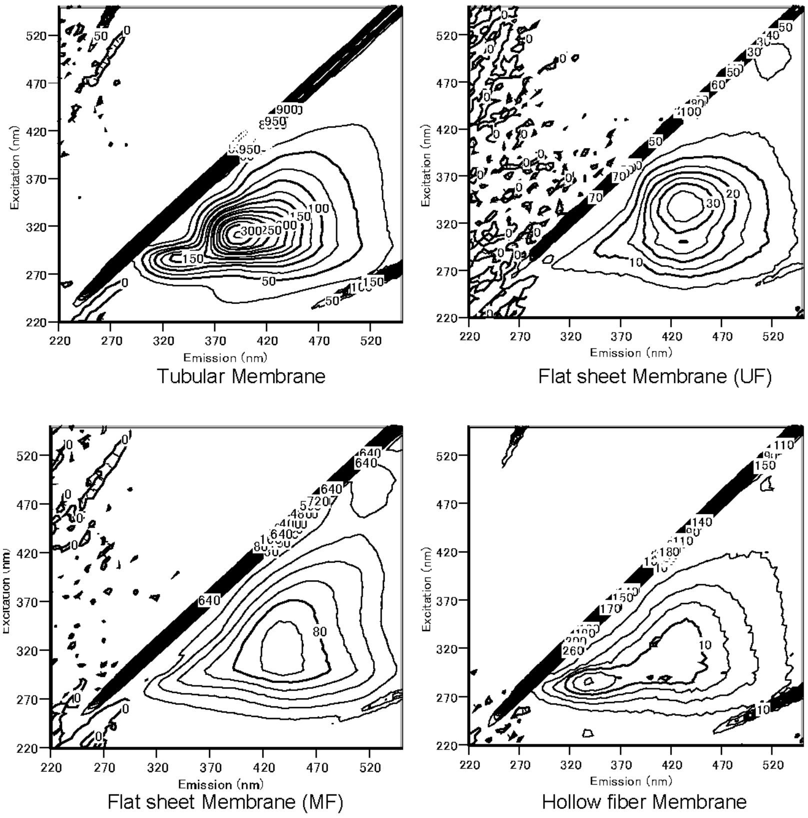

Fig. 5 (b). EEM Spectra of foulant in Run 2.

EEM spectra of foulant in Run 2 was presented in Figure 5.5(b). The same trend as that in Run 1 was seen in Run 2 for all types of membrane. The EEM spectrum measured for the foulant extracted from the hollow fiber membranes exhibited a different feature: a peak around 290 nm/ 330 nm (Ex/Em), assigned to protein-like substances, was the most intensive. Humic substances seemed to be less dominant in the foulant extracted from hollow-fiber membranes, in accordance with results of previous studies (Hoque et al. 2012).

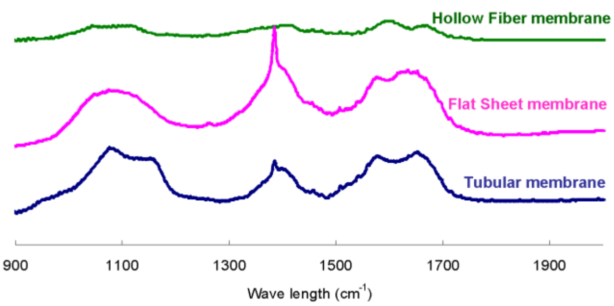

Fig. 6 (a). FTIR Spectra of foulant in Run 1.

The results of FTIR analysis also support the finding that humic substances were dominant in the foulants of flat sheet and tubular membranes. Figure 5.6(a), (b) shows FTIR spectra of foulants extracted from the fouled membranes in Run 1 and 2, respectively. A broad peak around 1400 cm-1 was significant in the spectra of the extracted foulant of tubular and flat sheet membrane, indicating the presence of symmetrical stretching of COO-, OH deformation and C-O stretching of phenolic group (Stevenson et al. 1971). These are the features found with humic substances. Peaks near 1600-1700 cm-1 specially found at 1660 cm-1 are indicators of amino groups (i.e. proteineous character) (Kimura et al. 2005; Barber et al. 2001), implying that proteins also contributed to membrane fouling to a minor extent. In contrast, intensities of peaks around 1050 cm-1, which are assigned to polysaccharides (Rosenberger et al. 2006).

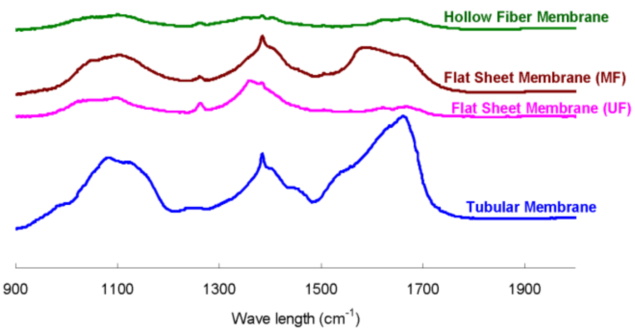

Fig. 6 (b). FTIR Spectra of foulant in Run 2.

3.3 Probable reason for the difference in different membrane

Although, in this experiment tubular, flat sheet (MF & UF) and hollow fiber membrane were examined using same mixed liquor suspension, an explicit comparison between these configurations has been done. In this study, the total resistance increase through the flat sheet membrane is significantly more rapid than through the tubular and hollow fiber membrane. All three types of membrane were PVDF membranes of different pore size. It would be also impossible to guarantee that all three types of membranes were identical material due to the specific chemical formulation; surface modification may differ from each other. Difference in chemical composition contributed to the difference in performance although the membranes were PVDF membranes.

This research demonstrated that flat sheet membranes (MF) membrane always foul more rapidly than hollow fiber and tubular membrane. Flat sheet (MF) membrane always fouled with in one weeks of operation. Cake layer were observed always in flat sheet membrane before physical cleaning. This cake layer might reduce the resistance. (Howe 2007) showed that flat sheet membrane fouled more rapidly than hollow fiber and tubular membrane. (Kurita 2015) also used granular material to reduce cake layer fouling in flat sheet membrane. In the case of tubular membrane, initial resistance was high compare to other two types of membranes. Characteristics of foulant also differed depending on membrane configuration. Humic substances show its dominance in flat sheet and tubular membrane whereas protein showed as dominant foulant in hollow fiber membrane.

4. Conclusion

A comparison between ASMBRs and submerged MBRs in terms of foulant characteristics was carried out. For this purpose, hollow fiber , flat sheet and tubular membranes made from the same polymer material (PVDF) were used both in side-stream and sub-merged MBR configuration. Membrane flux was set at 42 LMH for the three types of membranes and foulants were extracted from the membranes after 40 and 20 days of continuous operations in two Run for further analyses. The flat sheet (MF) membrane fouled within short period of time compared to other two types of membranes. Tubular membrane used as ASMBR was fouled the least among the three types of membranes. In the case of the characteristics of membrane foulant, there are some differences among different types of membranes. Humic like substance was found to be dominant in tubular and flat sheet membrane whereas, and protein like substance was found to be dominant in hollow fiber membrane.

References

Barber, L.B., Leenheer, J.A., Noyes, T.I., Stiles, E.A. (2001) . "Nature and transformation of dissolved organic matter in treatment wetlands." Environ. Sci. Technol. 35, 4805-4816.

Chen, W., Westerhoff, P., Leenheer, J.A., Booksh, K. (2003). „Fluorescence xcitation–emission matrix regional integration to quantify spectra for dissolved organic matter." Environ. Sci. Technol. 37, pp.5701–5710.

Dubois, M., Gilles, K.A., Hamilton, J.K., Rebers, P.A., Smith, F. (1956). "Colorimetric method for determination of sugars and related substances." Anal. Chem. 28, pp.350–356.

Hoque, A. Kimura, K., Miyoshi, T.,Yamato, N., Watanabe, Y. (2012). "Characteristics of foulants in air-sparged side-stream tubular membranes used in a municipal wastewater membrane bioreactor." Separation and Purification Technology 93 , pp. 83-91.

Howre, K.J., Marwah, A., Chiu, K.P., Adham, S.S (2007). "Effect of membrane configuration on bench-scale MF and UF fouling experiments." Water Research, 41, pp 3842-3849.

Judd, S. (2008). "The status of membrane bioreactor technology." Trends in Bio-technology. Vol 26 no 2, pp.109-116.

Kimura, K., Hane, Y.,Watanabe, Y., Amy, G., Ohkuma, N. (2004). "Irreversible membrane fouling during ultrafiltration of surface water." Water Res., 38, pp.3431–3441.

Kimura, K., Yamoto, N., Yamamura, H., Watanabe, Y. (2005). "Membrane fouling in pilot-scale membrane bioreactors (MBRs) treating municipal wastewater." Environ. Sci. Technol., 39, pp.6293–6299.

Kimura, K., Miyoshi, T., Naruse, T., Yamato, N., Ogyu, R, Watanabe, Y. (2008). "The difference in characteristics of foulants in submerged MBRs caused by the difference in the membrane flux." Desalination 231 . pp.268–275.

Kurita, T., Kimura, K., Watanabe, Y. (2015)."Energy saving in the operation of submerged MBRs by the insertion of baffles and the introduction of granular materials." Separation and Purification Technology. 141. pp.207-213.

Le-Clech, P., Jefferson, B., Judd, J., (2003). "Impact of aeration, solids concentrationand membrane characteristics on the hydraulic performance of a membrane bioreactor." J. Membr. Sci. 218, pp.117–129.

Le-Clech, P., Chen, V., Fane, T. A.G. (2006)."Fouling in membrane bioreactors used in wastewater treatment." J. Membr. Sci., 284, pp.17-53.

Lee, W., Kang, S., Shin, H., (2003)."Sludge characteristics and their contribution to microfiltration in submerged membrane bioreactors." J. Membr. Sci. 216,pp. 217–227.

Lowry, O.H., Rosebrough, N.J., Farr, A.L., Randall, R.J., (1951). "Protein measurement with the folin phenol reagent." J. Biol. Chem. 193, pp.265–275.

Rosenberger, S., Laabs, C., Lesjean, B., Gnirss, R., Amy, G., Jekel, M., Schrotter, J.C.(2006). "Impact of colloidal and soluble organic material on membrane performance in membrane bioreactors for municipal wastewater treatment." Water Res., 40, 710–720.

Stevenson, F.J., Goh, K.M. (1971). "Infrared spectra of humic acids and related substances." Geochimica et Cosmoschimica Acta, 35, pp. 471-483.