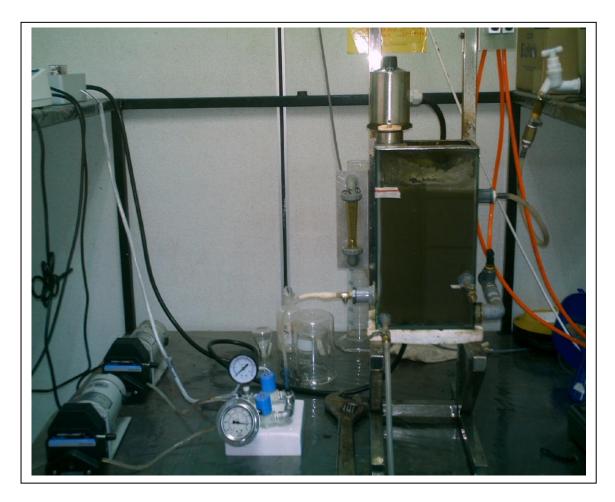
PROJECT PROFILE



The Performance of Submerged Membrane Bioreactor Treating High Temperature Wastewater

Research Area : Technology and Engineering

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ABSTRACT

Membrane bioreactor technology represents the most rapidly growing membrane technology in the water sector. In recent years, MBR technologies have been playing a very important role in water and wastewater treatment. MBRs are used to treat a wide range of municipal and industrial wastewaters. MBR integrates membrane filtration with biological degradation system of waste products. It is defined as a modification of the conventional activated sludge system (AS), where the separation of the treated water from the mixed liquor is accomplished by a membrane system instead of a clarifier. The two basic MBR process configurations are external and submerged. The high cost of pumping makes external MBR system impractical for full-scale municipal wastewater treatment plants. In this study, submerged MBR (SMBR) process was developed.

1. INTRODUCTION

1.1 Background

Membrane technology did not exist before the sixties of the last century (Richard, 2000). Furthermore, Christian (2005) has reported that in three decades, 50% of all separation processes will be accomplished by membranes. The first systematic studies of membrane phenomena are ascribed to the 18th-century philosophers and scientists, when Abbe Nolet in 1748 found the word *osmosis* to describe permeation of liquid through a diaphragm (Richard, 2000). The same researcher also reported that, through the 19th and early 20th centuries, membranes had no industrial or commercial applications, but they were used as laboratory tools to study physical and chemical theories.

Since 1960, interest in membrane filtration process has grown gradually, and membrane technology now is the object of substantial universal research, development, commercial activity and full-scale application (Joël *et. al.*, 1996). Hence, membrane filtration is on the edge of becoming a mainstream filtration process and is already competing with the conventional system techniques (Christian, 2005). Many researchers have defined membrane with different words. Joël *et. al.*, (1996) defined it as a thin layer of material that is capable of separation materials as a function of their physical and chemical properties when a driving force is applied across the membrane. Otherwise, membranes are often most of the times the first choice because of their decreasing costs, superior performance for improving a broad range of water qualities, use of less disinfection chemicals and smaller storage tanks and feed facilities (Christian, 2005).

Membrane filtration process has been utilized in a big range of applications. Membrane bioreactor (MBR) is one of them. MBR is a modification of the conventional activated sludge system (AS) using membrane instead of a clarifier to accomplish the process of separating treated water from the mixed liquor (Cicek *et. al.*, 1999). MBR technology combines the biological degradation process by AS with a direct solid-liquid separation by micro or ultrafiltration membrane technology (with a pore-size range of 0.05 to 0.4 μ m) (Pierre *et. al.*, 2006).

Unlike the conventional AS process which depends on a gravity settlement, MBR uses membrane filtration unit for the separation of biomass. Therefore, it is competent to complete biomass retention in the bioreactor and thus to retain potentially pathogenic organisms (Seung., 2004). In AS system, only the fraction of activated sludge that forms flocs and settles can be retained, while in MBR all components of the biomass that are larger than the membrane cutoff are retained. Thereby, MBR produces a high-quality and cell-free effluent, and reduces the need for disinfection necessities of treated wastewater effluents (Cote *et. al.*, 1998; Jefferson *et. al.*, 2000). Long SRT in the MBR process averts the washout of slow-

growing microorganisms such as nitrifying bacteria and other bacteria responsible for degrading complex compounds. Therefore, MBRs enhance the nitrifying function and complex organic contaminant degradation ability compared to a conventional biological wastewater process of AS system at short HRT (Muller et. al., 1995). Beside the superior effluent quality and the absolute control of solids retention and hydraulic retention times, the smaller volume and footprint is one of the main advantages of MBR.

MBR is an ideal option for municipal and industrial wastewater treatment applications, particularly in mesophilic condition. It has been exploited widely to treat various kinds of wastewater in many cities around the world. Nevertheless, MBR has not yet been utilized in the treatment applications of high-temperature (35 °C and above) municipal wastewater. This is more so in arid and dry-hot climate such as the Middle East consisting of countries i.e. Saudi Arabia, Oman, UAE, Qatar, Bahrain and Kuwait. In fact, there is a big usage of membranes for water and seawater treatment (desalination) in Gulf states, but not for wastewater treatment. Therefore, it is very important and necessary to study the feasibility of MBR in treating high-temperature municipal wastewater, especially, when there are no real studies on such subject.

Many researchers have been exploring the different applications of MBR process during last two decades. Majority of them focused on the performance of MBR at mesophilic conditions and low temperatures (Darren *et al.*, 2005; Aloice and Tatsuya, 1996; Zhang *et. al.*, 2006). Groups of researchers have studied the efficiency of MBR in treating various kinds of industrial wastewater, while other groups were involved in investigating the phenomena of membrane fouling (Ognier *et al.*, 2002; Pierre *et al.*, 2006; Fangang *et al.*, 2006). In spite of the efforts spent on studying the applications of MBR in treating high temperature industrial and synthetic wastewater (João *et. al.*, 2005 Zhang et al., 2005; Kurian & Nakhla, 2006;), the application of MBR in treating high temperature municipal wastewater remains very limited.

1.2 Aim and Objectives

Despite the big number of the previous studies related to the subject of MBR applications, the knowledge area of MBR treating HTMW has not yet been investigated before this study. The question of *"What is the effect of temperature on the performance of MBR process treating municipal wastewater"* has not yet been answered. Thus, the overall aim of this research was to study and evaluate the feasibility of MBR application process in treating high-temperature municipal wastewater, for the purpose of reuse and recycle. This can be achieved by the following specific objectives:-

I. To investigate the effect of high temperatures on the biodegradation process (biological removal efficiency) and membrane filterability (final removal efficiency) in MBR system treating municipal wastewater, in terms of COD, NH₃-N, SS, Turbidity and Effluent color.

- II. To investigate the effect of high temperatures on an activated sludge properties in terms of Biomass growth, SVI, Hydraulic viscosity, SMP and EPS ratio, pH and Supernatant turbidity.
- III. To investigate the phenomena of membrane fouling in terms of SMP and EPS ratio, and TMP and BWP.
- IV. To evaluate and compare between the performances of MBR process treating HTMW at two different (high and low) membrane hydraulic fluxes.
- V. To investigate the effect of drastic temperature changes on the performance of MBR process treating municipal wastewater, in terms of removal efficiencies, AS properties and membrane fouling phenomena at constant membrane hydraulic flux.

1.3 Scope of the study

Although significant work was conducted on MBR applications for high temperature wastewater treatment, there is a gap in literature in terms of the relationship between temperature and MBR process in municipal wastewater treatment. Many areas need to be investigated such as the relationship between the temperature and AS properties, biological removal efficiency, final removal efficiency, membrane fouling, and the effect of drastic temperature changes on the MBR process. Therefore, this research was initiated by conducting a thorough literature review on the use of MBR applications for different kinds of high temperature wastewater treatment, operational and performance factors influencing the process, types and efficiencies of removals, membrane fouling phenomena and biomass characterization. Based on this literature review, the gaps in knowledge and the needs to fulfill those gaps through this research have been established.

Based on the research objectives, the second task involved development and set up of appropriate lab-scale experiments and analytical methods to conduct the research. As far as the constraint and limitation of the study is concerned, the work is only enough for initial process development and membrane configuration. No analytical data could be established at this stage of study.

2. RESEARCH METHODOLOGY

2.1 Study perspective

Notwithstanding, the full-scale MBRs have been applied since the early 1970s (Ng, 2002), studies related to the submerged MBR started two decades ago upon the breakthrough for the MBR which came in 1989 with the idea of Yamamoto *et. al.* (1989), to submerge the membrane in the bioreactor. Until then, researchers had been interested in designing MBRs with the separation device located external to the reactor and relied on high trans-membrane pressure to maintain filtration. Many researches have been carried out to study the different sides of the MBR process and applications. MBR applications for treating different kinds of wastewater, operational modes and parameters, fouling phenomenon, designing and modeling have been investigated during the last ten years (Lesjean *et. al.*, 2004; Yaobo *et. al.*, 2006; Pierre *et. al.*, 2006; Anja *et. al.*, 2006; Sofia *et. al.*, 2004; Jan *et. al.*, 2007; Kim and Yuan., 2005; Nobuhiro *et. al.*, 2006).

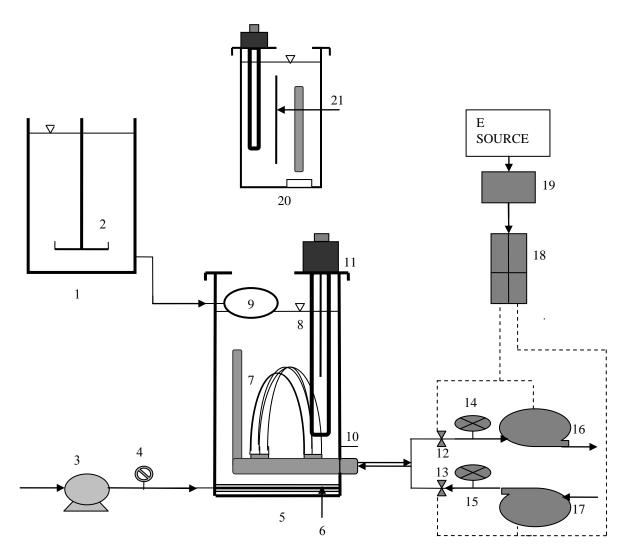
However, until now, there is still a lack of understanding in relation to performance of MBR in hot climate conditions for domestic wastewater treatment applications. This may be due to the fact that the origin of this technology was from the European continent. For hot and hightemperature climate regions like the Gulf States, MBR technology can still be considered as a new paradigm shift for wastewater treatment applications

2.2 Study outline

The experimental work for this study consisted of two parts. The first part is the system designing, modifying and examining, and the second part is the experimental runs which are explained in the next paragraph. However due to budget limitation, the study is confined to system designing and membrane configuration.

2.2.1 Bioreactor configuration

The MBR system and membrane modules were designed and modified by the researcher in the laboratory of Membrane Research Unit (MRU), Chemical Engineering Faculty, UTM Malaysia. This phase of system preparation (design, modification and examination) has taken nearly six months. It consisted of designing the bioreactor and membrane modules, and selecting the required materials according to the literature review. After that, the system was built, modified and examined prior to the commencement of the main experimental runs. The laboratory scale MBR system used in this study consisted of five main parts which are bioreactor, aeration system, heating system, membrane modules and suction/backwash set (Figures 2.1 and 2.2).



(1) Feeding tank (2) Mixer (3) Air compressor (4) Air flow meter (5) Main reactor (6) Aeration tubes with 1 ml openings (7) Membrane module (8) Water level (9) Water level controller (10) Sampling port (11) Thermocouple electric heater (12) Sucking automatic valve (13) Backwashing automatic valve (14) Suction pressure gauge (15) Backwash pressure gauge (16) Suction pump (17) Backwash pump (18) Programming time controller (19) Operational timer (20) Main reactor profile (21) Baffle plate

Figure 2.1 Schematic drawing of MBR system used in this study



Figure 2.2 Picture of MBR system used in this study

2.2.2 Main bioreactor

The bioreactor is made from glass with an effective volume of 3.6 L. The bioreactor's cover is made from steel and it contains a hole of 2 cm X 8 cm for the purpose of observation and temperature measurement. The baffle plate (15 cm X 18 cm) fixed in the middle of the bioreactor divided the bioreactor into two areas to keep the mixed liquor flowing up and down around the plate. The front area is the membrane submerging area and the back one is the heater submerging area. The water level controller is located 22 cm from the bottom in the right side of the bioreactor. The bioreactor contains one 2.5 cm hole at the bottom in the left side of the bioreactor for membrane module installation and suction/backwash tube fixing. It is situated 3 cm from the bottom of the bioreactor. The bioreactor contains also two 0.6 cm holes on the front side for sampling and desludging, and they are situated 6 cm and 1 cm from the bottom respectively. The effective volume of the bioreactor is 3.8 liters.

2.2.3 Aeration system

Aerobic condition was maintained by bubbling ambient air into the bioreactor at a rate of approximately 9 L/min measured using a hydraulic air flow meter. Aeration system consisted of compressor, air supply tubes, air flow meter and porous diffusing tubes. The compressor was self-operating and it's operating mode was approximately 3.5 min on and 15 min off. The 0.5 mm ID air supply tubes made of polyurethane (Sun-Rise Airkino) were connected from the compressor to the diffusing tubes through the air flow meter (Dwyer Instrument). The air flow meter was used to measure and control the air flow into the bioreactor. The three UPVC 65 mm ID diameter diffusing tubes (with many 1 mm pores) were positioned at the bottom of the bioreactor immediately below the membrane module so that the coarse bubbles would not only provide oxygen for the microorganisms, but also agitate the membrane fibers to reduce membrane fouling and prolong membrane service life.

2.2.4 Heating system (instrument)

The electrical heating system used in this study consisted of stainless steel heating electrode and temperature controller. The heating instrument was fixed on the steel cover of the bioreactor, whereas the heating electrode was submerged in the mixed liquor to raise up the bioreactor temperature. The temperature controller located outside the bioreactor was connected to a submerged sensor which switches the instrument on and of automatically to keep the temperature of the mixed liquor at a required constant-degree.

2.2.5 Membrane modules and specification

The membranes were ordered as a big bundle of commercial membranes, which were then modified into a laboratory scale modules. Different configurations of such modules have been modified and examined to select the suitable configuration for the experimental conditions of this study. Initially, various membrane configurations has been examined by using tap water and mixed liquor (sludge). Finally, the appropriate membrane module configuration was selected and used in the main stages of the experimental runs (Figure 2.3).

2.2.6 Suction/backwash set

The suction/backwash set is the part connected to the membrane module in the bioreactor. It consisted of two opposite-flow MasterFlex tubes (Cat no: 96410-16) for suction and backwash, two peristaltic MasterFlex pumps (Cole-Parmer Instrument Company, Model 77200-60) for suction and backwash, electrical timing controller to control the operating time of suction and backwash lines and electrical timer for operating and pause modes. There were two automatic valves between the membrane module and the suction and backwash lines. Each valve was opened when the line started working automatically. There was also a pressure gage between each valve together with pump on the same line.

2.2.7 Membrane cleaning chemicals and procedures

Since the hollow fiber membranes have relatively longer service life, the membrane modules used in this study were cleaned only at the end of each run or phase. After removing the membrane module from the bioreactor it was flushed under a running tap water to dislodge and remove the sludge suspended between the fibers. The module then was soaked in 0.5 % w/w NaOCI solution for 24 hours and then it was washed again with tap water and kept immersed in a sink of tap water until need. Sometimes the module needs to be soaked again in 0.5 % w/w NaOCI solution for 12 hours.



(a)



(b)

Figure 2.3 Samples of different configurations of membrane modules

The hollow fiber membranes used in this study were made of polyethersulfone which are hydrophilic and high temperature (≤ 100 °C), high pressure and different ranges of pH tolerant (Table 2.1).

Manufacturer	Aqueusphere
Membrane material	Modified Polyethersulfone (PES)
Nominal pore size (µm)	0.1-0.2
Fiber length (mm)	230
External diameter (mm)	0.9
Internal diameter (mm)	0.55
No of fibers in module	100
Total membrane surface area (m ²)	0.065
R m (m ⁻¹)	0.33 – 0.5
Operating Temperature	≤ 100 °C
Operating differentiate pressure	< 5 kPa
рН	2 – 11

 Table 2.1
 Membrane specifications

2.3 Operational parameters

2.3.1 pH control

The pH of the mixed liquor was continuously measured in order to be kept within the range of 7 \pm 1 by the addition of either a 1% HCL solution or a 20 g/L Na₂CO₃ solution and also the pH of influent and effluent. For pH measurement Thermo Orion pH Meter Model 420 was used.

2.3.2 Dissolved oxygen (DO)

DO was monitored using YSI 5100 DO meter. The membrane of the DO probe was replaced after four weeks to maintain the freshness. The DO probe was washed after each use and stored in a half water filled BOD bottle to maintain the moisture of the membrane.

2.3.3 Temperature (T)

Temperature was monitored continuously using Consort C535 Multi-Parameter analayser to measure influent, mixed liquor and effluent temperature.

2.3.4 Transmembrane pressure (TMP) and backwash pressure (BWP)

Both TMP and BWP were monitored by using two different pressure gages negative and positive respectively. The pressure gages are fixed on the suction and backwash lines between the peristaltic pumps and the submerged membrane module in the bioreactor.

3. LITERATURE REVIEW

3.1 Membrane Bioreactors

Membrane bioreactors (MBRs) are based on a combination of activated sludge processes and membrane filtration in one treatment step. An ultrafiltration or microfiltration membrane separates the activated sludge from the effluent. The membrane can be applied within the bioreactor (submerged configuration) or externally through recirculation. Since external settlers, or any other post treatment step, become superfluous by using a membrane for the suspended solid and effluent separation, the required space for an installation is small and sludge concentration in the aeration tanks can be two to three times higher than in conventional systems. Furthermore, the effluent quality is significantly better as all suspended and colloidal material such as micro contaminants, bacteria and viruses are removed (Ujang and Anderson, 2000; Trussell *et al.*, 2005).

Biological processes in a MBR are often comparable or better than in conventional activated sludge systems (Ujang *et. al.,* 2005 a, b and c). Due to the long sludge ages, N-removal is more efficient because the slow growing autotrophic bacteria are kept efficiently in the system. Denitrification can occur by introducing anoxic tanks or intermittent aeration (Drews *et. al.,* 2005; Gander *et. al.,* 2000). Figure 3.1 shows a typical MBR system.

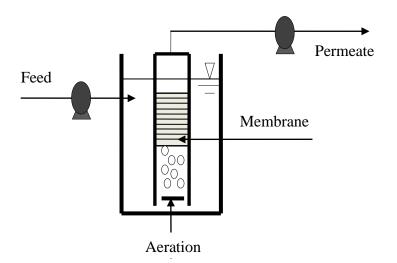


Figure 3.1 Typical membrane bioreator systme (Ujang and Anderson, 2000)

3.2 Membrane Bioreactor Technology and Process

3.2.1 Membrane Definition and Technology

Membrane basically, can be defined as a barrier, which separates two phases and restricts transport of various chemical in selective manner (Paul and Yampol, 1994). It can be also defined as a material that separates particles and molecules from liquids and gaseous. The membrane separation process is based on the presence of semi permeable membrane. The principle is quite simple: the membrane acts as a very specific filter that allows water to flow

through, while it catches suspended solids and other substances (Figure 3.2). There are two factors that determine the effectiveness of a membrane filtration process; selectivity and productivity. Selectivity is expressed as a parameter called retention or separation factor, while productivity is expressed as a parameter called flux.

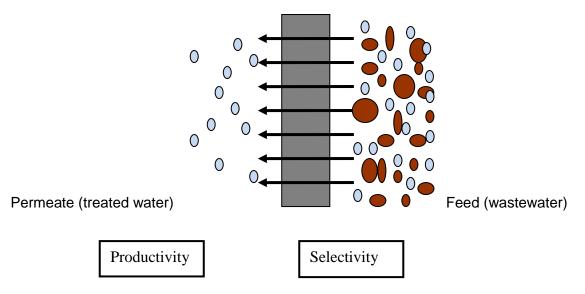


Figure 3.2 Schematic shape for membrane filtration process

3.2.2 Types of Membrane

There are many types of membrane modules used in MBR system according to the design and pore size. Membrane types according to the design are tubular, plate and frame, rotary disk and hollow fiber (Figure 3.3). The tubular membrane configuration is commonly used to enhance turbulent flow and mechanical cleaning. The plate and frame shaped membranes are inexpensive and usually disposable. The rotary disk membrane gives an acceptable membrane surface area. The module of hollow fiber is considered as a self-supporting membrane and presents the highest membrane surface area of all the membrane module types (Seung, 2004).

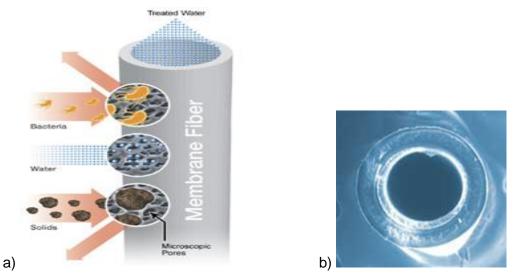
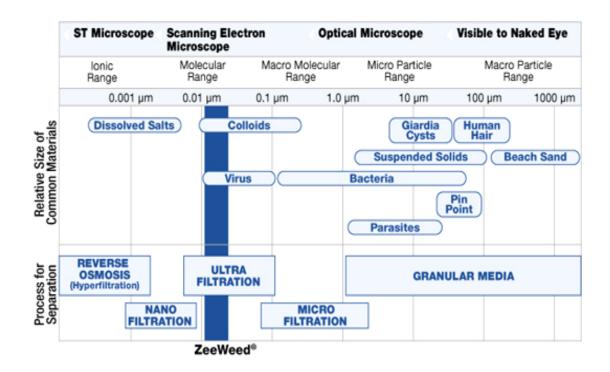


Figure 3.3 Hollow fiber membrane. a) fiber magnified several hundred times, b) a cross section of a membrane (Zenon, 2007)

Otherwise, there are four main types according to the pore sizes, which are Reverse Osmosis (RO), Nanofiltration (NF), Ultrafiltration (UF) and Microfiltration (MF). MF significantly removes particulate contaminants (clay, algae, bacteria and microorganisms) with minimal energy consumption among membrane family (Meier-Haack *et. al.*, 2003; Ujang *et. al.*, 2002). The range with pore size higher than that of MF is a granular media such as what is used in the conventional activated sludge system. For example, granular-sand filters (Table 3.1).

Table 3.1Membrane classification according to the pore size and retention capability(Zenon, 2007).



3.2.3 Membrane Bioreactor: Definition and Properties

As an alternative technology to the activated sludge process, researchers have been interested for about 35 years in integrating membranes with biological processes, to produce MBR system, which consists of bioreactor and membrane filtration unit (Christian, 2005). MBR for wastewater treatment is one of the fastest growing technologies in municipal and industrial wastewater treatment, especially for effluent reuse (Ng et al., 2004).

There are several drawbacks and disadvantages with the conventional system of municipal and domestic wastewater treatment. The production of sludge is high, nutrients could not be sufficiently removed and large footprint land areas required. Moreover, high numbers of filamentous bacteria causes bulking and severe solid-liquid separation problems, what reflects on effluent quality as a high concentration of suspended solid (Seung, 2004). However, MBR system can overcome all these problems and provide an improved effluent quality. MBR is a technology that will influence the future of wastewater treatment. MBR improves the quality of effluent by providing enhanced organic matter and nutrient removal, greatly reduces the quantity of solids discharged and remove pathogens, what eliminates the need for disinfection. All these advantages come with a smaller plant footprint, saving land costs (Dagmara *et. al.*, 2005). There are many advantages and properties associated with the MBR, which make it a reliable option over other treatment techniques such as:

- Small footprint requirement that reduces the cost of whole project (Dagmara *et. al.*, 2005; Judd, 2006)
- Compact system, thus easy to operate, monitor and maintain (Satoshi et. al., 2004)
- High effluent quality and good disinfection capability, which are often difficult to be effectively met by conventional activated sludge system (Judd, 2006; Zhang *et. al.*, 2006; Pierre *et. al.*, 2006)
- High capacity, which ranges from less than 1 m³/day to greater than 100,000 m³/day (Zhang *et. al.*, 2006)
- Complete physical retention of bacterial flocs, viruses, particulate matters and all suspended solid within the bioreactor (Pierre *et. al.*, 2006; Chiemchaisri *et. al.*, 1992)
- Ability to accumulate and successfully operate with relatively high mixed liquor suspended solids (MLSS) concentration (up to 30 g/L), which allows long sludge retention time (SRT; up to 200 days) in moderately sized bioreactor (Davies *et. al.*, 1998; Darren *et. al.*, 2006)
- High volumetric loading and low-sludge production, which reduces the cost of sludge disposal (Cicek, 2003; Judd, 2006)
- High nitrification activity rates owing to small floc size of the biomass (Zhang *et. al.*, 1997; Cicek *et. al.*, 1999)
- Eliminate process problems and difficulties associated with settling, which are the most troublesome part of wastewater treatment (Cicek, 2003)
- Low-pressure system, thus reduces energy and operating cost (Sandeep et. al., 2002)
- SImple to be controlled and modeled (Ng et. al., 2004)
- Relatively, simple cleaning materials and procedures, thus reduces maintenance cost (Ujang, 2000b)
- Becoming cheaper over the years (Stephen et. al., 2004)

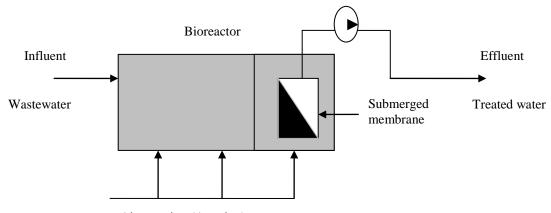
3.3 MBR Types

According to its position, MBR system can be classified into two major groups: internal (submerged) and external (sidestream or recirculated). Based on the electron acceptor for the biological reaction, the MBR system can be classified into two groups: aerobic and anaerobic MBR (Seung, 2004).

3.3.1 Internal and External

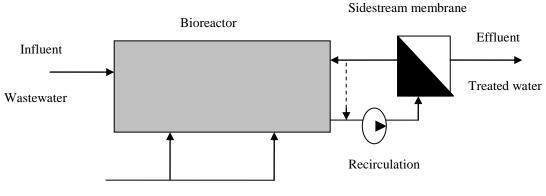
The first group is the internal (submerged) MBR, in which membrane filtration unit is integrated into biological reactor to treat and separate biomass (Engelhardt *et. al.*, 1998). Recently, this type of MBR has become a promising alternative to the conventional treatment, thus it has been developed to simplify the system and reduce the operational cost (Darren *et. al.*, 2006). The driving force across the membrane in the submerged MBR is achieved by creating a negative pressure on the permeate side of the membrane unit (Yamamoto *et. al.*, 1989 and Chiemchaisri *et. al.*, 1993). The second group, is the external (sidestream) MBR, in which, the mixed liquor is recirculated through a membrane filtration unit. The driving force in sidestream MBR, is the pressure obtained by high cross flow velocity through the membrane filtration unit (Winnen *et. al.*, 1996 and Urbain *et. al.*, 1996). Although, the high cost of mixed liquor recirculation in sidestream MBR, higher effluent fluxes, easier maintenance and less complicated configuration make it desirable (Seung, 2004). Figure 3.4 (Paul *et. al.*, 2006), simply shows the two types of MBR.





Air sparging (Aeration)





Air sparging (Aeration)

Figure 3.4Simplified schematics of MBR configurations (internal and external)(Paul *et. al.*, 2006)

For moderate to large-scale municipal wastewater treatment, submerged systems are preferred over sidestream configuration, due to small footprint and reactor requirements. Even though the submerged type is younger than other, approximately 55% of MBR installations are in submerged type while the remainder are in the sidestream type (Stephenson *et. al.*, 2000).

3.3.2 Aerobic MBR

The aerobic MBR has been applied quite widely to domestic, municipal wastewater treatment instead of the conventional activated sludge system (Gander *et. al.*, 2000; Jefferson *et. al.*, 2000; Ueda and Hata, 1999 and Murakami *et. al.*, 2000). Darren *et. al.*, (2005) reported that, their laboratory-scale aerobic MBR system managed to remove 98% of the suspended solid and achieving a remarkable COD removal efficiency of 96% in treating high strength synthetic wastewater. However, phosphorus removal in MBR varied from 12% (Cote *et. al.*, 1997) to 74% (Ueda and Hata, 1999). The concentration of the MLSS is reported to be 10 g/l and up to 50 g/l in some studies (Muller *et. al.*, 1995 and Scholz and Fuchs, 2000).

Aerobic MBR has been applied to treat a wide range of industrial wastewater, such as oily (Scholz and Fuch, 2000 and Seo *et. al.*, 1997) and tannery wastewaters (Yamanoto and Win, 1991). Despite the high strength of the industrial wastewater, many studies have reported high COD removal efficiency with high organic loading rate (Scholz and Fuch, 2000; Yamanoto and Win, 1991; Kurian and Nakhla, 2006 and Rozich and Bordacs, 2002). Aerobic biological process operated at high temperatures is highly advantageous in treating high temperature, high strength industrial wastewaters due to its ability to integrate the advantages of conventional aerobic and anaerobic processes that include rapid biodegradation kinetics and low biological solids production respectively (Rozich and Bordacs, 2002). The low yield of 0.03 g VSS/g COD, observed by Kurian and Nakhla (2006) reveals that the aerobic MBR is a potential solution to difficulties related to high sludge generation in conventional systems treating high strength wastewaters.

3.3.3 Anaerobic MBR

Although, the disadvantages of anaerobic MBR such as lower growth rate, high MLSS concentration requirement and long HRT to prevent the biomass from washout, there are advantages of the anaerobic MBR over the aerobic one, which are biogas recovery, lower sludge production and lower energy consumption regarding to the absence of aeration process (Seung, 2004). Although, anaerobic MBR to date is less explored than aerobic MBR, it is a promising system for different strength wastewater treatment with simultaneous energy recovery and less excess sludge production (Dongen *et. al.*, 2004). Due to the poor settleability in gravity settlers and the strong potential for odors, the use of anaerobic process previously was unfeasible for nutrient and COD removal in municipal wastewater (Seung, 2004). Thus, many researchers have been studying the anaerobic MBR in the application of

high strength industrial wastewater treatment (Choo *et. al.*, 1996). COD removal efficiency in anaerobic MBR was 97% at high loading rate and >90% at low loading rate (Strohwald and Ross, 1992). However, the theoretical range of biogas production is 65-75%, while Strohwald and Ross (1992) in treating brewery wastewater reported the removal of between 20 and 47% mass of feed COD. 50% of biogas produced from industrial wastewater was found to be methane content according to Kang *et. al.*, (2002) and it could be more than that depending on the type of the treatment application.

3.4 MBR Performance and Operating Factors

This section describes the performance of MBR, such as in terms of sludge production, COD, nitrogen and phosphorus, and the factors affecting the permeate flux rate.

3.4.1 Sludge Production

Low sludge production was observed in the MBR processes because of limited energy source (Witzig *et. al.*, 2002), mechanical shear caused by pumping (Choo and Lee, 1996; Kang *et. al.*, 2002; Kim *et. al.*, 2001), or attachment on to the surface of membrane (Choo and Lee, 1996). In addition, the sludge age also influences the biomass production. Chaize and Huyard (1991) demonstrated that sludge production was greatly reduced if the sludge age is between 50 and 100 days. The performance of MBR process has been shown to be satisfactory for at least two months when the sludge is completely retained (Chiemchaisri *et. al.*, 1993). However, it is unclear if the accumulation of inert material has a negative effect on the treatment performance. The ratio of VSS to total suspended solids (TSS) in MBR MLSS was reported in the range of 0.46 - 0.55 (Seung, 2004), which is much lower than the 0.75 - 0.90 observed in activated sludge MLSS.

3.4.2 Removal Efficiency

a. COD Removal

The MBR system is capable of achieving COD removal by both physical and biological mechanisms. The biological COD removal occurs in the bioreactor. The biological COD removal efficiency can be calculated from the difference of soluble CODs in the feed and the mixed liquor divided by soluble COD in the feed (Ng *et. al.*, 2000). The membrane filter offers the physical barrier against particulates and some soluble organic carbon and inert fractions of the mixed liquor (Chang *et. al.*, 2001). The biological COD removal increases with time, but the physical COD removal by membrane decreases over time because of the age of the membrane and sloughing of some biomass on permeate side of the membrane (membrane fouling). Chang *et. al.* (2000) proposed the mechanisms of COD removal by membrane to be due to three mechanisms; sieving method depending on membrane pore size and cut-off, adsorption into membrane pores and surface, and sieving and/or adsorption onto the cake layer.

The COD concentration also can be reduced by gas production under anaerobic conditions (Anderson *et. al.*, 1986; Choo and Lee, 1996; Kang *et. al.*, 2002). Strohwald and Ross (1992) also showed that about half of influent COD was converted to methane gas and the ratio increased slightly with the increase of HRT. Sufficient biomass can ensure good performance in COD removal and better quality effluent. If enough biomass concentration is present, the increased TSS concentration in the bioreactor does not significantly affect COD removal.

The changes in HRT and SRT do not significantly influence the COD removal in the MBR. However, Darren *et. al.* (2006) observed slight difference in overall COD removal efficiencies of MBRs treating the same wastewater at different HRTs. Overall COD removals were 97.63%, 96.88% and 96.54% at 24h, 12h and 6h respectively. In previous studies, some researchers reported that the filtration membranes in submerged MBR were more severely fouled at high sludge concentration (Magara and Itoh, 1991; Manen and Sanderson 1996), while others suggested that higher sludge concentration resulted in less fouling under certain conditions (Defrance and Jaffrin 1999; Lee *et. al.*, 2001).

The effect of high temperature on the removal efficiencies in MBR was studied by Zhang *et. al.* (2006). The removal efficiency was more than 97% at 35 and 40 °C, while it was 93% at 45 °C. The same researchers reported that the richness in microbial diversity reduces in high temperature treatment because of the sudden changes in operational conditions. This microbial diversity decay could cause lower removal of pollutants (Tripathi and Grant.1999; LaPara *et. al.*, 2000). In all previous studies, mesophilic activated sludge processes have produced higher COD removal than thermophilic processes (Zhang *et. al.*, 2006).

b. Nitrogen Removal

Nitrification

Nitrification is the conversion to nitrate by microorganisms. This is achieved in two stages with the conversion from ammonia to nitrite by *Nitrosomonas* followed by the conversion from nitrite to nitrate by *Nitrobacter* or *Nitrospira*. The nitrite concentrations do not build up in most biological treatment systems because *Nitrobacter* or *Nitrospira* immediately converts this compound to nitrate. Both nitrifying organisms are autotrophs which use inorganic carbon (carbon dioxide, bicarbonate, or carbonate) for cell synthesis, and ammonia or nitrite to derive energy. They grow slower than most of the heterotrophic microorganisms. Fan *et. al.* (1996) found 0.1 to 0.2 d⁻¹ of nitrifier growth rates in an MBR treating municipal wastewater. As nitrifiers are slower in their growth than heterotrophs, longer sludge ages are required in order to achieve full nitrification (>90%). The high nitrification can be observed in the aerobic MBR because membrane separation entirely confines the nitrifying bacteria within the bioreactor independent of sludge concentration. In addition, as sludge production is low in MBR, nitrifying bacteria face less competition from heterotrophic bacteria which also consume ammonia. Cote *et. al.* (1997) reported that ammonia removal efficiency was

improved by increasing the sludge age from 10 days to 50 days. Xing *et. al.* (2001) observed a high nitrification rate at 3.75 hours of HRT and 5 days of SRT.

There are three substrates for completing nitrification, which are carbon dioxide, ammonia and oxygen. Nitrification is inhibited at low DO level (< 1 mg/L), but it is completely recovered again after DO level increase (Chiemchaisri *et. al.*, 1992). Nitrification is inhibited due to high free ammonium concentration (> 8.4 mg/L) and high DO demand (Ng., 2000). The nitrification has been reported to be increased with increase in temperature up to approximately 30 °C, and slowed down as the temperature increases beyond that. At low temperature, nitrification is more severely affected than denitrification due to temperature changes (Fdz-Polanco *et. al.*, 1994).

Denitrification

Denitrification is the reduction of the oxidized nitrogen to N_2 gas (Yamamoto *et. al.*, 1989; Chiemchaisri *et. al.*, 1992). This process requires a suitable electron donor, which is usually an organic compound. The optimal pH for denitrification is neutral to slightly alkaline (Metcalf and Eddy., 1991). The reduction of nitrate to nitrogen gas by the denitrification produces alkalinity, resulting in elevated pH.

Denitrification is one of the most efficient methods for removal of excessive amounts of nitrates in the wastewater. Denitrifying bacteria are mostly facultative anaerobes and heterotrophs. In order to achieve denitrification, intermittent aeration mode, which gives anoxic conditions, can be applied to aeration tank in an MBR system without the deterioration of permeate quality (Chiemchaisri *et. al.,* 1992; Ueda *et. al.,* 1996).

Traditionally, nitrification and denitrification are used for nitrogen removal from wastewater. However, these processes may not be energy effective because these processes require aeration for oxidizing ammonia to nitrate (nitrification) and COD for reducing nitrate to nitrogen gas (denitrification). In addition, major denitrification gas product can be N₂O, which is known as a very strong greenhouse, not N₂ at the higher nitrite concentration (Zeng *et. al.,* 2003).

c. Phosphorus Removal

The enhanced biological phosphorus removal can be obtained from the selective enrichment of bacteria accumulating inorganic polyphosphate with a cyclic regime of alternating anaerobic and aerobic condition (Zhao *et. al.,* 1994). Under anaerobic condition, the intracellular poly- β -hyroxy-alkanoates (PHA) are formed from the substrate and the stored polyphosphate is hydrolyzed to soluble orthophosphate. Under aerobic condition, the stored PHA is consumed for growth and maintenance of the cell as well as the uptake of the soluble orthophosphate. Biological phosphorus removal is usually integrated with biological nitrogen removal in wastewater treatment.

Intermittent aeration can achieve nitrogen and phosphorus removal by enhancing the process of simultaneous nitrification and denitrification (SND), phosphorous uptake and phosphorus release in the same reactor. However, even though intermittent aeration was successful in removing nitrogen, phosphorus removal was difficult to achieve at the same time. It was probably due to incomplete denitrification (Seung. 2004).

In MBR systems, phosphorus removal ranged from 11.9 % to 75 %. Using the intermittent aeration submerged MBR, Seo *et. al.*, (1997) obtained 66 % phosphorous removal. However, the filtration operation was limited to during the aeration period only in the intermittently aerated and submerged MBRs. Therefore, a continuous aerated MBR with a separated anoxic tank can improve the phosphorus removal efficiency. Cho *et. al.*, (2003) reported 93 % phosphorus removal with a sequencing anoxic/anaerobic membrane bioreactor, since it's removal under the continuous aeration is rather low.

3.4.3 Membrane Fouling in MBR

MBR technology represents the most rapidly growing membrane technology in the water sector, with an estimated global market of US\$ 216.6 million in 2005 rising at annual growth rate of 10.9% and an expected value of US\$ 363 million in 2010 (Hanft, 2006). However, as for all membrane processes, MBR is ultimately restricted by the tendency of the membrane for fouling, which causes a reduction in permeability and demands frequent physical and chemical membrane cleaning (Guglielmi et. al., 2007). All factors affecting the performance of MBR could contribute in producing the phenomena of membrane fouling. Thus, membrane fouling is an ideal application to study the factors affecting the performance of the MBR process.

Koros *et. al.*, (1996), has defined the term of membrane fouling as *"the process resulting in loss of performance of a membrane due to the deposition of suspended or dissolved substances on its external surface, at its pores openings or within its pores"*, what results as an increasing in transmembrane pressure TMP.

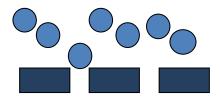
Membrane Fouling

Particle separation and water permeation involve various mass transport steps in membrane filtration processes. Mass transfer can be limited by the attachment, accumulation or adsorption of materials on the membrane surface and/or within membrane pores. As a result, increase in hydraulic resistance over time is expected. This phenomenon is called membrane fouling (Zhou et. al., 2001). Fouling have various origins as classified here (Mulder, 2000; Duranceau, 2001). Biofouling: adhesion and accumulation of microorganisms, biopolymers

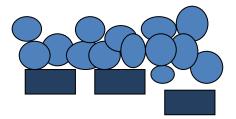
(Extracellular polymeric substances and soluble microbial products), Bacterial fouling; Particle and colloidal fouling: deposition of clay, particles humic substances, debris and silica; Crystalline fouling (scaling): deposition of mineral due to excess of the solutes.

Fouling affects the performance of the membrane either by deposition of a layer onto the membrane surface or by blockage or partial blockage of the pored (Field *et. al.*, 1995). Three fouling mechanisms were introduced for membrane filtration in general that can be applied for MBR as well (Knyazkova *et. al.*, 1999) (Figure 3.5):

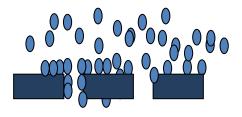
- Pre closure or pore narrowing: when diameter of particles is smaller that diameter of pores, particles could enter the pores. As a result some of the entered particles pass the membrane and some foul inside the pores and reduce the open cross-sectional area for flow.
- Pore plugging: for the case when diameters of particles are similar to those of the pores, particles block the pores.
- Cake formation: for the case when diameters of particles are bigger than diameter of pores, particles deposit on the membrane surface. This leads to cake build-up (cake formation).



a) Pore Plugging



b) Cake Formation



c) Pore Closure

Figure 3.5 Three main mechanisms for membrane fouling

Fouling inside membrane pores by salt precipitates and small colloids is considered an irreversible process. On the other hand, flux decline due to the development of cake on the membrane surface is largely reversible. The indicator of this form of fouling is long-term decline in the flux rate.

3.4.4 Fouling Characteristics and Mechanisms

Biofouling

Biofilm is a matrix of cells and cellular products, such as biopolymers (EPS and SMP), attached to a solid surface of membrane (Hardorfer *et. al.*, 1999). Mostly, microorganisms are growing as sessile communities and this could be due to the protective nature of biofilm growth (Walker *et. al.*, 2000). In MBR processes, a high concentration of microorganisms is used to biodegrade the nutrients in the wastewater, as a result MBR processes provide a good environment for biofouling formation (Flemming, 2000).

Most of the studies on macromolecular fouling (with biological origin), have been based on protein and carbohydrate fouling which are the major components of biopolymers (EPS and SMP) found in MBR. In number of studies fouling was evaluated by protein and carbohydrate in fouling layers and permeate solution (Ji and Zhou., 2006; Zhang *et. al.*, 2006; Janga *et. al.*, 2007). Presence of both protein and carbohydrates around the biological cells was proposed as a key parameter in the flocs formation and it may have a significant role in MBR fouling (Gorner *et. al.*, 2003). It was observed that by changing the concentration of protein from 20 to100 mg/L, the specific resistance value increased by a factor of 10.

During protein filtration, different fouling phenomena can be expected including protein adsorption, deposition and mass-transfer limitation due to concentration polarization or boundary layer effects. Protein adsorption is a specific interaction between proteins and membrane polymer that could occur in the absence of convective flow through the membrane (Bowen et. al., 1991). Protein deposition refers to any additional protein that fouls on the membrane surface during filtration. It is ultimately forms on the upper surface of the membrane as a cake formation. This layer could be very effective on flux decline; two orders of magnitude decline in the permeate flux as a result of protein deposition is dependent on pH value (Palecek *et. al.*, 1994). Protein deposition was a minimum at the protein iso-electric point and decreased with the increase in ionic strength. It was also found that by reducing the electrostatic repulsion between proteins, more compact deposit layer would be expected. Protein denaturation and/or aggregation is one of the most widely suggested mechanisms in the initial stage of protein fouling during membrane microfiltration (Kelly *et. al.*, 1993).

Direct relationships between the carbohydrate level in soluble microbial products SMP solution and fouling rate was observed in filtration of municipal wastewater (Lesjean *et. al.*, 2005). Fouling in submerged membrane bioreactors (MBRs) was studied for flux stepping filtration on seven biomasses under different operating conditions (Germain *et. al.*, 2005). Similar result was observed recently by Janga, *et. al.* (2007), where concentration of carbohydrate was reported as the highest in comparison with other foulants in the feed solution. Another study was carried out on SMPs in MBR at different sludge retention time (SRT) (Liang *et. al.*, 2007). Protein and carbohydrate were reported as components of SMP. However it was shown that hydrophilic neutrals such as carbohydrates were the main foulants of SMP. Similar results were was claimed in MBR (Wang et. al., 2006). In this research statistical analysis was used intensively, and carbohydrate found to have significant influence on membrane fouling.

Several researches were carried out by using sodium alginate as a model solution for carbohydrate to be able to analyze the effect of carbohydrate individually on fouling. Alginate was chosen as a model EPS and fouling mechanism was studied during the dead end unstirred microfiltration of sodium alginate (Ye *et. al.*, 2005). It was observed that the fouling layer formed in the long term subcritical flux operation appeared to be irreversible, while fouling layers formed in the short term dead end constant pressure or flux stepping tend to be more reversible. Combination of pore blockage and cake formation was proposed as a possible fouling mechanism.

Bacterial fouling is the other form of biofouling which is considered as a problem in UF and MF systems. The surface of bacteria cells consist of a peptidoglycan layer covalently linked with different membrane proteins and anionic polymers. Irreversible adhesion of one or more bacteria to the membrane surface, initiate the membrane biofouling. It is followed by growth and multiplication of the sessile cells at the expense of feed water nutrients (Ridgway *et. al.*, 1999).

The formation of biofouling and biofilm on the membranes can be divided into three steps: the initial step is the development of conditioning layer. This layer is caused by the adsorption of macromolecules, organic acids and lipopolysaccharides on the membrane surface. Attachment of microorganisms to this conditioning film is the second step; this step appeared to occur rapidly. The rate is highly dependent on the concentration of microorganism, type and nutritional status of the microorganisms in the process water. The third step is the colonization and multiplication of the microorganisms on the membrane surface and formation of irreversible blocking of the membrane (Kabsch-Korbutowicz, 1992).

Bacteria use different complex strategies to adhere to different surfaces. For example, bacterial fimbriae, flagella or fibrils are long filamentous projections of the cell surface that

can act as bridging structures to overcome repulsive electrostatic interactions between negatively charged bacteria and negatively charged surfaces (Zeman *et. al.*, 1996). Membrane surface charge is an important factor in some selected biofouling , but it is not applicable for all kinds of biofoulings (Bharwada *et. al.*, 2000).

Once microorganisms are bound to the membrane surface, they can grow and use the nutrients in the process stream. Feed stream with high levels of total organic carbon cause a severe biofouling. Adherent bacteria produce a variety of exopolysaccharides, which then become part of extracellular slime or biofilm on the membrane surface. It is believed that this complex layer is acting as an ion-exchange resin for enhanced nutrition. This layer also improves the long term aggregation and adhesion of these bacteria. The bacteria within the biofilm are more resistant to bacterial agents. As a result, it may be difficult to remove this layer by physical or chemical cleaning methods (Zeman et. al., 1996).

It was shown that the biofilm consisting of variety of bacterial types can provide a different hydraulic resistance to the permeate flux (Hodgson *et. al.*, 1993). There is a very close relationship between MBR process conditions and microorganism distribution. It was shown that the biofouling on the membrane is depending upon membrane operating condition as well as the properties of the activated sludge in the MBR systems (Choi *et. al.*, 2006). The biofouling phenomenon in suspended and attached growth MBR was evaluated (Sombatsompop *et. al.*, 2006). It was observed that increase in fouling was associated with increasing MLSS concentration.

Particle and Colloidal Fouling

The contents in MBR filtration feed solution is mostly particles such as microorganism cells, inorganic and organic particles. These particles are retained by membrane and generally formed a cake layer on the membrane surface. This cake layer decreases the hydraulic permeability of the membrane. The characteristics of the cake layer are dependent on the operating conditions and the property of the particulates.

Crystalline Fouling (Scaling)

Mineral material deposition on the membrane surface is called crystalline fouling which is due to high concentration or charge interaction between membrane and filtrate ions. It is the most common form of fouling in desalination plants. However, due to high biological fouling in MBR, crystalline fouling has not been taken into consideration and this factor was rarely reported in the literature.

3.4.5 Factors Affecting MBR Fouling

The three main factors affecting the rate of fouling in MBR will be detailed in the following sections and include (Hillis, 2000):

- Membrane properties (membrane material, pore size and distribution and module configuration).
- Mixed liquor characteristics (nature and concentration of the bulk fluid such as MLSS concentration, EPS and particle size distribution).
- Operating conditions (factors such as permeate flux, cross flow velocity or aeration intensity, hydraulic retention time (HRT), and SRT).

3.5 Membrane Properties

3.5.1 Hydrophobicity

Hydrophilicity is related to the chemical characteristics of the membrane which allows the material around the membrane, to wet the membrane and form a water film or coating on their surface. Hydrophobic materials have little or no tendency to adsorb water, and water tends to stay on their surface. On the other hand, by increasing the hydrophilicity of a material, the association between water, through hydrogen bonding, and hydrophilic material increases. For microfiltration (MF) membranes, hydrophobicity influence wettability, applied pressure requirements for liquid flow through the membrane and fouling propensity (Meng *et. al.*, 2006a).

Hydrophobicity of the surface of membranes influences fouling. Many natural products, due to dipole or multiple chemical bonds in their structure, are negatively charged while particulate foulants in aqueous media are more generally hydrophobic. As a result, particles attach to any material less hydrophilic than water. By attachment of the particles to the membrane surface, less exposure of hydrophobic particles can be achieved. More fouling was observed during the hydrophobic membrane filtration in comparison with the hydrophilic one (Belfort *et. al.*, 1994; Chang *et. al.*, 1999; Judd and Till., 2000; Choi *et. al.*, 2002). For wastewater treatment, the membrane should preferably be hydrophilic (Fane *et. al.*, 1991).

3.5.2 Membrane Charge

The potential electric field created by this charge can attract or repel charged species in water. Since the natural organic macromolecules in water and wastewater are generally negatively charged, the negative or neutral membrane are preferred in order to limit the particle adsorption (Cardew and Lee., 1998).

3.5.3 Membrane Pore Size

In practical, the type of membrane filtration is chosen according to particles size distribution and purpose of filtration. During the MF and UF, membrane filtration was compared (Palecek *et. al.*, 1994). It was found that UF membranes are less prone to fouling by macromolecules, since the smaller pores are more impenetrable. It was shown that, initial fouling is expected on larger pore size membranes as a result of pore blocking (Hong *et. al.*, 2002). It has been demonstrated that MF has a higher permeate flux than UF membrane at the same condition due to larger pore size in the former situation. The permeate flux for MF declines much quicker than UF. The performance of nanofiltration MBR in domestic wastewater treatment was examined (Choi *et. al.*, 2007). By analyzing the molecular weight of dissolved organic matter from nanofiltration MBR, it was shown that the pore size increased over operating time. The influence of membrane pore size modification on membrane performance was investigated and It was shown that the flux can be increased and decreased by stretching membranes (Worrel *et. al.*, 2007).

Limited number of studies has been done on the optimal membrane pore size for wastewater treatment. The pore size membranes for the current commercial submerged MBR processes are approximately 0.04-0.4µm.

3.6 Mixed Liquor Characteristics

3.6.1 Mixed Liquor Suspended Solid (MLSS) Concentration

In the development of MBR technology, many studies focused on the effect of MLSS concentration on the membrane fouling and controversial findings about the effect of this parameter have been reported. The MLSS concentration in MBRs is not a dominant factor influencing the overall membrane fouling unlike the dead-end membrane filtration (Chang *et. al.*, 2005). It is reported that membrane fouling took place more rapidly at high MLSS concentrations (Chang *et. al.*, 2002). Exponential relationship between MLSS concentration and membrane fouling resistance was reported (Meng *et. al.*, 2006b). On the other hand, some authors have reported positive impact of MLSS (Defrance and Jaffrin., 1999). Also some researchers reported insignificant impact of MLSS on fouling behavior (Hong *et. al.*, 2002; Le-Clech *et. al.*, 2003; Lesjean *et. al.*, 2004; Lesjean *et. al.*, 2005). At MLSS concentrations of higher than 30,000 mg/L, no significant effect of MLSS on irreversible fouling was observed (Lubbecke *et. al.*, 1995). All these experiments were carried out on different range of MLSS concentration.

3.6.2 Viscosity

In both processes, conventional activated sludge and MBR, biomass viscosity is closely related to its concentration and has been reported as a foulant parameter (Yeom *et. al.*, 2004). A critical MLSS concentration is the point under which the viscosity rises slowly and increases exponentially above (Itonaga *et. al.*, 2004). The significance of MLSS viscosity is that it modifies bubble size, inhibits hollow fibers movement in submerged bundles and reduces the efficiency of oxygen mass transfer (Wicaksana *et. al.*, 2006 and Germain and Stephenson. 2005). Therefore, the main result of high biomass viscosity would be high membrane fouling rate.

3.6.3 Extracellular Polymeric Substances (EPS)

Extracellular polymeric substances (EPS) have been identified as the main foulants in membrane bioreactor (MBR) operation (Rosenberger *et. al.*, 2003; Janga *et. al.*, 2007). EPS are high molecular-weight mucous secretions from microbial cells. EPS matrix is very heterogeneous and can be characterized by its relative levels of polysaccharides, proteins, and more rarely lipids and nucleic acids (Nuengjamnong *et. al.*, 2005; Janga *et. al.*, 2007). For membrane units filtering activated sludge, biofouling remains a major issue as organic adsorption and deposition on membrane surface significantly reduce hydraulic performances, leading to rise in operational and maintenance costs.

EPS are produced by most bacteria and participate in the formation of microbial aggregates whether the bacteria grow in suspended culture or in biofilms (Flemming *et. al.*, 2001). EPS are mainly responsible for the structural and functional integrity of biofilms and are considered as the key components that determine the physicochemical and biological properties of biofilms. In general, the proportion of EPS in biofilms can vary between roughly 50 and 90% of the total organic matter (Nielsen *et. al.*, 1997). They consist of insoluble materials such as sheaths capsular polymers, condensed gels, loosely bound polymers and attached organic material; these are produced by active secretion, shedding of cell surface material or cell lysis (Janga *et. al.*, 2005).

Chang and Lee (1998) measured the EPS content quantitatively by separating the activated sludge broth into three portions, i.e., cell, bulk, and EPS fraction. It was found that EPS was the major contributing component to the total fouling resistance. EPS matrixes are multiple and they include aggregation of bacterial cells in flocs and biofilms. They make a protective barrier around the bacteria, as a result retention of water and adhesion to surface is expected (Laspidou and Rittmann., 2002). EPS can form a highly hydrated gel matrix that microbial cells are embedded in, due to its heterogeneous and changing nature (Nielson and Jahn., 1999). Therefore the EPS content of activated sludge was suggested as one of the probable index for the membrane fouling in an activated sludge MBR system. In addition, bioflocs attached to the membrane can be very effective in MBR by playing as a nutrient source during the biofilm formation on the membrane surface (Ishiguro *et. al.*, 1994; Flemming *et. al.*, 1997).

EPS can be classified as extracted (eEPS) which are artificially produced from the biological cell floc and the soluble EPS which are present in the activated sludge supernatant and are not associated with the cell (soluble microbial products or SMP) (Le-Clech *et. al.*, 2006). The term "EPS" is used as a general parameter to characterize the biopolymers in the reactor (Figure 3.6). So far no standard method for extraction exists, during the studies on the effects of EPS in MBR fouling; as a result it is difficult to make a comparison between research groups. Due to the simplicity, the heating method is sometimes preferred to extract the eEPS

(Figure 3.7). The eEPS is then characterized in terms of protein (eEPSp) and carbohydrate contents (eEPSc) by using colorimetric methods: Lowry (Lowry *et. al.*, 1951) and Dubois (Dubois et. al., 1956) protocols respectively. The eEPS solution also can be characterized in terms of total organic carbon (TOC) (Cho *et. al.*, 2005; Nagaoka and Nemoto., 2005). It was observed that eEPSp is more hydrophobic and eEPSc is more hydrophilic (Liu and Fang., 2003). By comparing the results in literature, higher level of eEPSp was generally reported in comparison to that of eEPSc (Le-Clech *et. al.*, 2006).

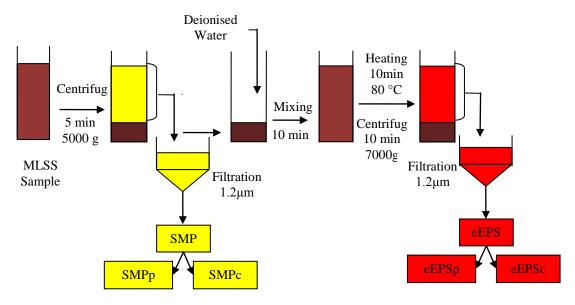
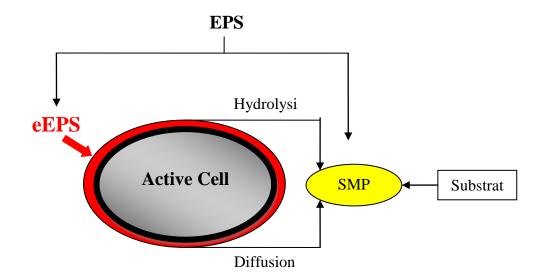
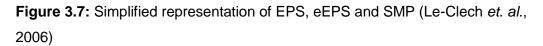


Figure 3.6: Heating method for EPS and SMP extraction and measurement (Le-Clech *et. al.*, 2006)





3.6.4 Soluble Microbial Products (SMP)

SMP are defined as soluble cellular components that are released during cell lysis, diffuse through the cell membrane, are lost during synthesis (Laspidou and Rittmann., 2002; Li et.

al., 2005). In MBR systems, they can also be provided from the feed substance. During filtration, adsorption of SMP on the membrane surface and blocking of the membrane pores are expected; it leads to the formation of gel structure on the membrane surface where they possibly provide a nutrient source for biofilm formation and a hydraulic resistance to permeate flow (Rosenberger *et. al.*, 2005). Four MBR case study based on SMP analysis was also studied; in order to show the feasibility and relevance of liquid phase analyses on MBR filterability and potentially standardize the method.

In order to separate the water phase from the biomass, three different methods were investigated. It was found that simple filtration through the filter paper is the most effective method in comparison with centrifugation and sedimentation (Evenblij and van der Graaf., 2004).

Similar to eEPS, SMP solution is characterized with its protein and carbohydrate contents (Evenblij and van der Graaf., 2004), with its TOC level (Gao *et. al.*, 2004) or rarely with SUVA measurement (Shin and Kang., 2003). It is observed that there is no significant change in SMP characterization during a weekly measurement from the same reactor; also in terms of molecular weight distribution, SMP feature larger macromolecules (Brookes *et. al.*, 2003). Similar analysis was carried out on submerged MBR (Janga *et. al.*, 2007). It was observed that most of the SMPp in the reactor existed at a MW above 10 kDa and over 86% of SMPc contain in the permeate, had a MW below 1kDa.

Direct linear relationships between loss of MBR hydraulic performances and SMP concentration have been reported for an anaerobic MBR, (Fawehinmi *et. al.*, 2004a). It was observed that for MBR sludge level of EPS was unchanged while the SMP components could be accounted for higher membrane fouling (Cabassud *et. al.*, 2004). During this study, biological activities were observed which was indicating the presence of free bacteria in the MBR supernatant. It could also be another reason for membrane fouling. Creation of fouling layer on the membrane surface would act as a second membrane that can increase the adsorption of macromolecules and/or the retention (Rosenberger *et. al.*, 2006). As the permeate flows through the membrane, the formation of a biofilm could also assist in the degradation of macromolecules; interaction between the macromolecules and other solutes such as humics and divalent cations within the membrane pores may be the explanation for reduction of membrane pore size over time.

According to the results presenting the direct relationship between the carbohydrate contents in SMP solution with fouling rate (Lesjean *et. al.*, 2005), filtration index and CST (Evenblij *et. al.*, 2005), critical flux (Le-Clech *et. al.*, 2005) and specific flux (Rosenberger *et. al.*, 2005), SMPc can be revealed as the major foulant in MBR systems.

3.6.5 Particle Size

For MBR systems, the sludge particle size is around 20-40 μ m (Zhang *et. al.*, 1997). In this range of particles, shear-induced diffusion dominates the particle back-transport (Belfort *et. al.*, 1994). It was shown that shear-induced hydrodynamic diffusivity is positively proportional to the square of the particle diameter multiplied by shear rate (Eckstein *et. al.*, 1977). Therefore, easier detachment of particle with a larger size from the membrane surface is expected.

3.6.6 Temperature

The effect of temperature on membrane filtration process affects the permeate fluid viscosity (Mulder, 2000). For comparing the hydraulic performance obtained at different temperatures, normalization of the operating flux at reference temperature (25°C) is commonly used.

Effect of temperature was investigated during the filtration of municipal wastewater through MBR pilot plant (Jiang *et. al.*, 2005). Two sets of temperatures (17-18°C and 13-14°C) were used. The higher resistances were observed at lower temperature and it was explained by four following phenomena occurred in the system: (1) the viscosity of sludge was calculated within that temperature range, and it was increased for 10% as a result of reducing the shear stress generated by coarse bubbles, (2) building up of deflocculating tend to happen at low temperature, releasing EPS to the solution and reducing biomass floc size, (3) particle back transport velocity which was calculated with the Brownian diffusion coefficient (linearly related to temperature, biodegradation of COD was also decreased; resulting in higher concentration of particle COD and solute in the reactor (Jiang *et. al.*, 2005). The last phenomenon was also reported in other research with higher SMP which was measured in an anaerobic MBR and operated at 20°C (Fawehinmi *et. al.*, 2004b). Since all of these factors are directly linked to membrane fouling, greater deposition of materials on the membrane surface at lower temperatures is expected (Rosenberger *et. al.*, 2006).

3.6.7 pH

The influence of pH on membrane fouling has been widely investigated, since pH influences the electrostatic interaction between particles and particles-membrane. MF of Bovine serum albumin (BSA) and lysozyme solutions were carried out at different pH values (Ouammou *et. al.*, 2006). After analyzing the results in terms of blocking filtration laws and substantial changed in the fouling behavior, it was observed that fouling was a function of the solution pH.

3.7 Design and Operating Conditions

Several design and operation parameters including the shear rate, the imposed flux and SRT are likely to influence the membrane fouling of MBR processes:

3.7.1 Shear Rate /Aeration

High shear rates generated at the membrane surface, remove deposited material and as a result it reduces the hydraulic resistance of the fouling layer. With the high concentration of activated sludge within the bioreactor, high intensity scouring of the membrane is necessary to maintain permeate flux rates. Air scouring or circulation flow generate a crossflow velocity, which directly affect the cake layer formation over the membrane (Chang *et. al.*, 2000). Both experimental and empirical studies have revealed the influence of cross-flow velocity or aeration on membrane fouling (Bouhabila *et. al.*, 1998). The effect of aeration on cake removal and TMP in submerged MBR pilot plant was examined (Ueda *et. al.*, 1997). It was shown that fouling was reduced at higher air flow rate conditions. When hollow fibers were used fouling limitation by aeration was also due to agitation of the membrane (Wicaksana *et. al.*, 2005). For reducing membrane fouling it is suggested to increase the air flow rate or increase the aeration intensity by concentrating on membrane modules over a small floor area (Wicaksana *et. al.*, 2005).

3.7.2 Critical Flux and Determination Methods

Permeate flux determines the fouling rate (Zeman *et. al.*, 1996). It is generally found that membrane fouling increases with increasing flux (Judd, 2004). However it has been reported that fouling is not observed when the flux is retained below the critical flux (Chang *et. al.*, 2002).

Initially, the concept of critical flux was introduced by Field *et. al.* (1995). According to this hypothesis, the critical flux is the maximum flux, below which a decline of flux with time does not occur. Another definition is that it is a point at which fouling become irreversible (Howell, 2004). This concept has been applied for systems involving macromolecules, colloids, particles, bacteria and biomass.

The lateral migration theory has been used to explain the flux-paradox phenomena for colloidal suspension and it was proposed that when the lift velocity (VL) at the colloidal cake surface equals or exceeds the oppositely-directed membrane permeation velocity (J), fouling would not be expected to occur (Green and Belfort., 1980). Based on this theory it was mentioned that if J is less than VL, particle would not deposit on the membrane surface (Fig.3.8 (a)) (Kwon *et. al.*, 1998). This situation is below critical flux. On the other hand, the deposition of particles appears when J exceeds VL (Fig.3.8 (b)).

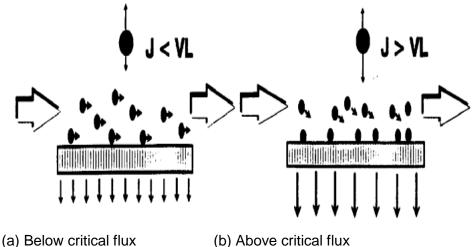


Figure 3.8: Different circumstances of critical flux in microfiltration.

There are two forms of critical flux: strong and weak. The strong form states that the subcritical flux-TMP relationship shows a straight line of the same slope as that of pure water for the same operating pressure, while the weak form shows a straight line, the slope of which differs from that of pure water. Any deviation from the straight line for either form indicates above critical flux conditions.

The concept of critical flux has been applied to systems involving macromolecules, colloids, particulates, bacteria and biomass. In order to indicate the fouling, in each flux step, flux was increased and decreased while TMP was recorded; the difference between TMPs called deviation. If deviation equaled to zero, it was presumed that no fouling had occurred. The point, in which deviation increases, is called critical flux.

The measurement of particle mass balance is another method in which the loss of mass in a bulk liquid above the membrane is measured by detecting concentration differences in the bulk fluid. The maximum flux at which no particle deposit on a membrane occurred (no difference in concentration of feed) can be determined as a critical flux (Kwon *et. al.,* 2000).

3.7.3 Sludge Retention Time (SRT)

2001). It was found that physicochemical properties of sludge surfaces, such as hydrophobicity and surface charge are influenced by the SRT (Liao *et. al.*, 2001). It was also established that at higher SRT, less negatively charged and more hydrophobic are expected than at lower SRT.

3.8 Limitation and cleaning procedures

The cleaning procedures used to remove membrane fouling when it occurs are two types: physical and chemical. However, there are other methods that could be used to prevent fouling before it occurs in what is known as fouling limitation.

3.8.1 Limitation

The major techniques for extending the life of membrane as long as possible before fouling are three. These techniques are (Pierre *et. al.*, 2006):

- Optimizing the anti-fouling properties and characteristics of membrane by increasing the membrane hydrophilicity and addition of TiO2 nanoparticles to the casting solution of membrane.
- Optimizing the operating condition of the MBR by injecting air to the reactor (aeration), operating MBR at low fluxes and modifying SRT and reactor design.
- Modifying the biomass characteristics by adding ferric chloride, aluminum and adsorbents.

3.8.2 Cleaning procedures

Physical cleaning

Physical cleaning techniques for fouling removal in MBR process are membrane relaxation and backwashing. The membrane relaxation is a pause of filtration for a short while after a period of suction (for example 10 min suction followed by 30 sec pause). The backwashing (backflushing) is a pumping of permeate again in reverse to the reactor through the membrane (for example 10 min suction followed by 30 sec backwashing) (Pierre *et. al.*, 2006).

Chemical cleaning

In addition to the physical cleaning techniques, chemical cleaning is also recommended. The prevalent chemical cleaning agents used in the normal condition are sodium hypochlorite (for organic foulants) and citric acid (for inorganics). These chemical cleaning techniques include (Pierre *et. al.*, 2006):

- Chemically enhanced backwash (daily), to intensify the efficiency of the backwash.
- Maintenance with higher chemical concentrations (weekly), to maintain the permeability and decrease the frequency of the intensive cleaning.
- Intensive chemical cleaning (once or twice a year), to recover the filtration ability of the membrane.

3.9 High-temperature treatment (thermophilic treatment)

Thermophilic reactor coupled with membrane (thermophilic MBR) is not a new concept and it has been experimented by Lopetegui and Sancho (2003) and Klatt and Lapara (2003), by using an immersed membrane to treat high-temperatures wastewater.

High-temperature treatment is considered to be feasible for wastewaters with high oil and grease content, owing to the strong solubility of this contaminant at higher temperatures. Despite this advantage in thermophilic treatment, many researchers (Çetin and Sürücü, 1990; Barr *et. al.*, 1996; Tripathi and Allen, 1999 and Lapara and Alleman, 1999) have reported the deterioration of sludge settleability with raising temperature. This inability of sludge separation from effluent liquid combined with low sludge yields could result as a biomass washout and low quality effluent. However, membrane technology was successfully employed to overcome this problem.

Thermophilic treatment is an attractive alternative for industries producing hot process waters and wastewaters. Operation under thermophilic conditions can be very useful for aerobic industrial wastewater treatment systems in different ways (Zhang, *et. al.*, 2005). High COD removal efficiency and low sludge net yield are the obvious advantages of thermophilic treatment (Couillard and Zhu, 1993; Tardif and Hall, 1997). Moreover, membrane could suffer from fouling problems at low temperatures than high temperatures (Jiang *et. al.*, 2005). Otherwise, the combination of the thermophilic aerobic process and a membrane bioreactor effectively solves the problem of solid-liquid separation. The thermophilic MBR process has high biomass concentrations and high loading rates, and it produces good-quality effluent (Ramaekers *et. al.*, 2001 and Huuhilo, *et. al.*, 2002). Tardif and Hall (1997) had compared different alternatives for recirculated newsprint whitewater treatment and found MBR to be the most reliable under high temperatures.

4. FINDINGS

4.1 Introduction

This chapter discussed the results obtained from the experimental works during the five stages of start-up, 25 °C, 35 °C, 45 °C and drastic temperature changes. The chapter is divided into three main headings, which are start-up, membrane fouling phenomenon and removal efficiency. The discussion is done by analyzing the results using different computer programs such as Excel and Statistic Plus. Comparison between the results obtained was also carried out to determine the differences between the parameters under varying conditions.

4.2 Start-up

After accomplishing the stage of system design and set-up, this stage of start-up was began. In this stage the performances of the membrane bioreactor system were investigated. The connections between the reactors were checked and any malfunctions in the design and installation of the reactors were rectified. It included also, selection of the perfect membrane module configuration after examining four of them. Moreover, some necessary tests such Critical Flux, K_{CW} , K_{Sludge} , TMP_{CW} , TMP_{Sludge} and dP/dt were determined. The system was operated by using tap water for one week and then by using municipal AS brought from Muscat City for more than two months to acclimatize and to allow bacterial growth to occur in the reactor.

4.2.1 Membrane Module Configurations

Four membrane module configurations were designed and modified by the researcher in the laboratory of Membrane Research Unit (MRU) in UTM. These membrane module configurations were installed in the system and examined by using tap water each for one day to compare between their performances and select the optimal. Among the four configurations two were selected to be examined using real sludge, where the better one would be selected. The parameters used in the selection process were flow stability, air bubble suction, fibers swaying mode and membrane fouling (Table 4.1).

Parameter	Description
Flow stability	Continuity of identical water quantity drawn out from the system (effluent flow) during a certain period of time, measured in L/min
Air bubble suction	Draw of air scored by bubble course during suction process, what reduces system flow out amount (effluent quantity)
Swaying	Fibers movement in swaying motions due to air bubbles
Membrane fouling	Membrane pores clogging causing increasing TMP

Table 4.1 Parameters used for selection process of membrane module configurations

Module configuration 1

This configuration has the suction line located on the top right (Figure 4.1). The performance of this module was quite satisfactory but resulting in substantial increase in TMP which was not justified. Although the medium used was tap water, the flow was steady without air in the out flow and with normal fibers swing, but the TMP was rather higher than the expected mode. The TMP started with 107 mbar at 10 LMH and increased with the time. This relatively high TMP could lead to inaccurate readings which may reflect negatively on the efficiency of the study. Hence, this configuration was eliminated.

Module configuration 2

A drop shape module configuration with the open fiber edges gathered at one end and connected to the suction line (Figure 4.2). The fibers in this configuration were located immediately after the aeration tubes and were freely swaying. The fibers in this configuration were very proximate to the bubbles source (aeration tubes). Therefore, it sucked the air beside the water what impacted the system out flow and increased the TMP. For these reasons this configuration was also eliminated.

Module configuration 3

A fixed top, multiple bundle membrane module configuration with a left bottom suction line (Figure 4.3). Performance of the module was excellent in a medium of tap water. The flow was smooth and steady without air in the out flow and the fibers swaying was relatively limited since the top of the module was fixed. Due to the smooth flow, this configuration was one of the two configurations selected to be applied in activated sludge medium. It was later eliminated due to early membrane fouling as a result of rapid increased in TMP. The rapid membrane fouling was caused by sludge attachment within the fibers due to their limited swaying movements.

Module configuration 4

A module similar to module configuration 1 but with a suction area at the left bottom (Figure 4.4). The module performance was the best in both tap water and sludge mediums. The flow was very smooth in the tap water medium and was steady in the sludge medium. There was no sucked air in the out flow and the fibers motion was more free. This configuration exhibited reasonable increase in TMP, which was 20 mbar and 100 mbar at 10 LMH and 15 LMH respectively with low fouling rates during a period of five days for each flux. This configuration was the most workable and suitable, thus it was selected to be used in this study.



Figure 4.1 Membrane module configuration 1



Figure 4.2 Membrane module configuration 2



Figure 4.3 Membrane module configuration 3



Figure 4.4 Membrane module configuration 4

After membrane configuration has been established, the study was continued without any grant available.

5. CONCLUSION

In this study, a laboratory scale membrane bioreactor system was designed, fabricated and employed for treating high temperature municipal wastewater at two different hydraulic fluxes. The study was mainly carried out to investigate the effect of temperature changes on the performance of MBR in treating municipal wastewater at low and high hydraulic fluxes. However the experimental works could not be carried out to completion, since only the membrane configuration was established. Further works were carried out without any grant allocation. Therefore, the conclusions that can be drawn were based on membrane configuration, and the recommendations for the future works were listed.

5.1 Conclusions

The conclusions that can be summarized were as follows:

Start up stage

- i. Membrane module configuration with a suction area in the left bottom was the most workable and suitable. Thus it was selected to be used in this study. The module performance was the best in both mediums of tap water and sludge. It's flow was very smooth in the tap water medium and it was steady in the sludge medium. There was no sucked air in the out flow and the fibers motion was more free. This configuration exhibited reasonable increase in TMP, which was 20 mbar and 100 mbar at 10 LMH and 15 LMH respectively with low fouling rates during a period of five days for each flux.
- ii. TMP increased gently until the flux of 12 LMH was achieved. Then, it ascended significantly beyond the flux of 14 LMH until the peak point. In clean water, TMP was zero up to the flux of 11 LMH and then increased gradually to 0.062 bar at 24 LMH. While, in a sludge medium it was zero up to 8 LMH then increased linearly up to 14 LMH. Above a flux value of 14 LMH, a distinct break occurred in the curve with a substantial change in TMP beyond flux of 16 LMH. With in the area of 14 to 16 LMH, the curves of flux-permeability and flux-fouling rate crossed each other. Thus, the critical flux is within the range of 14 to 16 LMH or presumably is 15 LMH, an interval corresponding to the results indicated by Defrance and Jafferin (1999), Ognier *et. al.* (2004) and Yang *et. al.* (2006).

5.2 Recommendations

i. By using the same system that was used in this study, a study can be carried out to compare between the performances of different membrane modules (hollow fiber, flat sheet and stainless steal) in treating municipal wastewater at high temperatures.

ii. By modifying the system that was used in this study via adding anaerobic reactor before the aerobic one, a study can be carried out to investigate the P Bio and Fin removal efficiencies. The study also can extend the period of the stages to estimate the acclimatization of the MBR system with the new conditions on the long term. The periods can be eight weeks instead of four. The obtained results then can be compared with the results obtained by this study.

6. RESEARCH OUTPUT

6.1 Citation Details of Articles
6.2 Citation Details of Conference Papers
6.3 Citation Details of Other Publications - books / standards etc. (*Please specify*)
6.4 Details of IPR (*Please specify*)

Due to the incompletion of study i.e. for only one year duration with allocation of RM10,000, the research output is only the establishment of membrane configuration and bioreactor design. No published papers could be made at the end of the study.

7. HUMAN CAPITAL DEVELOPMENT

7.1 Details of Human Capital Development (Name and qualification sought)

One postgraduate student at PhD level is envisaged. However after a one year period, the study was continued without any funding.

8. AWARDS / ACHIEVEMENT

8.1 Details of Recognition Received No available recognition received.

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