REMOVAL OF FORMALDEHYDE IN SYNTHETIC WASTEWATER USING SBR AND MBR

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หัวข้อวิทยานิพนธ์	การกำจัดฟอร์มัลดีไฮด์ในน้ำเสียสังเคราะห์ด้วยระบบเอสบีอาร์และเอ็มบีอาร์
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บทคัดย่อ

งานวิจัยนี้ศึกษาการบำบัดน้ำเสียสังเคราะห์ที่ปนเปื้อนฟอร์มัลดีไฮด์โดยใช้ระบบบำบัดแบบเอสบีอาร์ และเอ็มบีอาร์ การทดลองแบ่งออกเป็น 3 ขั้นตอน คือ ขั้นตอนที่ 1 การศึกษาหาสภาวะที่เหมาะสมโดยทำการทดลองแบบแบทช์ ปัจจัยที่ศึกษาได้แก่ ระยะเวลาสัมผัส ความเข้มข้นของฟอร์มัลดีไฮด์ ปริมาณตะกอนจุลินทรีย์ และค่าพีเอช ขั้นตอนที่ 2 ศึกษาประสิทธิภาพการบำบัดฟอร์มัลดีไฮด์ด้วยระบบเอสบีอาร์ และขั้นตอนที่ 3 ศึกษาประสิทธิภาพการบำบัดฟอร์มัลดีไฮด์ด้วยระบบเอ็มบีอาร์ จากการศึกษาพบว่า ระบบบำบัดทางชีวภาพแบบเติมอากาศสามารถบำบัดฟอร์มัลดีไฮด์ที่มีความเข้มข้นสูงถึง 1,000 มก./ล. ได้ร้อยละ 99.6 ที่เวลา 24 ชั่วโมง และ ร้อยละ 99.9 ที่เวลา 48 ชั่วโมง โดยปริมาณของตะกอนจุลินทรีย์ เท่ากับ 500 มก./ล. การเพิ่มปริมาณตะกอนจุลินทรีย์ส่งผลให้เพิ่มประสิทธิภาพการบำบัด โดยระยะเวลาบำบัดสั้นลง พบว่าปริมาณตะกอนจุลินทรีย์ในช่วง 1,000–2,000 มก./ล. สามารถบำบัดฟอร์มัลดีไฮด์ได้ร้อยละ 99.9 ที่ระยะเวลาสัมผัสตั้งแต่ 6 ชั่วโมงขึ้นไป น้ำที่ผ่านการบำบัดมีค่าฟอร์มัลดีไฮด์ประมาณ 1 มก./ล. การศึกษาผลของพีเอช พบว่า ค่าพีเอชที่เหมาะสมอยู่ระหว่าง 5-7 การบำบัดด้วยระบบเอสบีอาร์สามารถบำบัดฟอร์มัลดีไฮด์ที่มีความเข้มข้น 500 มก./ล. โดยค่าประสิทธิภาพการบำบัดเฉลี่ยร้อยละ 99.7 โดยอายุสลัดจ์ที่ 60 วันให้ค่าประสิทธิภาพการบำบัดฟอร์มัลดีไฮด์สูงกว่าที่ 30 และ 10 วัน การบำบัดฟอร์มัลดีไฮด์ด้วยระบบเอ็มบีอาร์พบว่า ประสิทธิภาพการบำบัดฟอร์มัลดีไฮด์มีค่าเฉลี่ยร้อยละ 99.7 ซึ่งไม่แตกต่างกับระบบเอสบีอาร์ แต่ข้อดีของระบบ เอ็มบีอาร์คือ น้ำทิ้งมีความใสโดยของแข็งแขวนลอยถูกกำจัดร้อยละ 100 อายุสลัดจ์สูงให้ประสิทธิภาพการบำบัดมากกว่า และให้ค่าฟลักซ์คืนกลับหลังการทำความสะอาดเยื่อกรองมากกว่าที่อายุสลัดจ์น้อย

คำสำคัญ : การอุดตัน, ฟอร์มัลดีไฮด์, เยื่อกรอง, อายุสลัดจ์, เอสบีอาร์, เอ็มบีอาร์

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ABSTRACT

This research studied the treatment of formaldehyde in synthetic wastewater using sequencing batch reactor (SBR) compared with membrane bioreactor (MBR). The experiments were divided into 3 stages: 1) Batch tests: to study factors affecting biological processes and optimum conditions, including contact time, formaldehyde concentration, mixed liquor suspended solids (MLSS), and pH, 2) Sequencing batch reactor (SBR), and 3) Membrane bioreactor (MBR). The results showed that aerated biological process treated formaldehyde at a concentration up to 1,000 mg/L with percent removal of 99.6 and 99.9 at a contact time of 24 and 48 hours, respectively, with MLSS of 500 mg/L. An increase of MLSS enhanced the efficiency of formaldehyde removal at shorter period of contact time. The MLSS in a range of 1,000 – 2,000 mg/L was found to remove 99.9% of formaldehyde at a contact time at least 6 hours. The formaldehyde concentration in effluent was approximately 1 mg/L. The optimum pH was in between 5 and 7. The SBR and MBR were operated with optimum conditions from batch tests. The SBR could treat formaldehyde at concentration of approximately 500 mg/L with average removal efficiency of 99.7%. Better performance of the SBR was achieved at solid retention time (SRT) of 60 days than at the SRT of 30 and 10 days. The MBR was effective in formaldehyde removal, with average percent removal of 99.7%, similar to the SBR. The MBR provided other benefits in terms of TSS removal and system stability. The effluent from the MBR was transparent with TSS removal of 100%. Longer SRT provided higher efficiency on formaldehyde removal efficiency and flux recovery after cleaning than shorter SRT.

Keywords : Fouling, Formaldehyde, Membrane, Sludge age, SBR, MBR

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Kamolchanok Sangsawang

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CHAPTER 1

INTRODUCTION

1.1 Research motivation

Formaldehyde is a compound that commonly used in large variety of processes in chemical industry, such as formalin, polyester fiber, adhesives, synthetic material, and resin production (Kaszycki *et al.*, 2001; Campos *et al.*, 2002). Because of its high toxicity, formaldehyde is often used as an active ingredient in preservatives and disinfectant agents to inhibit microbial activity (Eiroa *et al.*, 2005). It is frequently found in wastewater and waste gas causing environmental pollution. Formaldehyde present in industrial wastewaters can be found at concentrations up to 10,000 mg/L (Zoutberg and de Been, 1997). Formaldehyde is classified as a very toxic agent with severe negative action on living organisms. According to national industrial effluent standard, formaldehyde concentration of more than 1 mg/L is prohibited to discharge into receiving water, therefore, industries have to provide appropriate treatment methods to remove formaldehyde in wastewater before discharging into the rivers.

Treatment of formaldehyde in industrial wastewater can be accomplished by several treatment methods, including physical, chemical, and biological processes. Adsorption by activated carbon was found to remove formaldehyde from wastewater, however, it exhibited low efficiency (Tanada *et al*, 1999). Oxidation processes using some oxidizing agents, e.g. ozone (O_3), hydrogenperoxide (H_2O_2), and chlorine (Cl_2), could remove formaldehyde efficiently, but they have some limitations regarding to operation and maintenance cost, as well as the occurrence of some residuals of toxic substances. Biological process is found to be a good way to treat formaldehyde in industrial wastewater because it is an economic and reliable system for the treatment of wastewater. Activated sludge process is the most commonly used technology for biological wastewater treatment. It consists of a biochemical stage (aeration tank) for the degradation of contaminants by activated sludge and physical stage (secondary clarifier) for biomass separation and recirculation. Sequencing batch reactor (SBR) is a modified type of activated sludge processes with a combination aeration and clarifier in the same tank. Typically, a cycle of SBR operation consists of fill, react, settle, and draw steps. However, in case the toxic compounds for the microorganisms are present in the wastewater, that biological treatment

methods can be situated in trouble, leading to low efficiency in formaldehyde removal. Although, formaldehyde is a toxic compound for most microorganisms, some researchers found that specific strains of microorganisms e.g. *Pseudomonas* spp. generum, *Halomonas* spp., various strains of methylotropha ; *Debariomyces* spp. and *Trichosporon* spp. yeast genera, *Hansenula* spp., *Candida* spp., and *Gliocladium* spp. fungi can live in condition of high formaldehyde concentration (Glancer-Soljan *et al.*, 2001). Nevertheless, conventional activated sludge process with these specific microorganisms can treat formaldehyde, some of them may be washed out of the bioreactor due to their settle ability. This situation leads to instability, or even a failure of biological system in the treatment of formaldehyde. If these specific microorganisms can be stored in the system, the treatment processes would be operated consistently. Membrane bioreactor (MBR) is an interesting alternative method for wastewater treatment because it combines the activated sludge process with a membrane separation unit, instead of clarifiers (Matosic *et al.*, 2006). Membrane bioreactor provides more advantages than the conventional activated sludge process in terms of low sludge production and less area requirement, in addition microbial population in membrane bioreactor is well acclimated and capable of degrading specific substrates, i.e. formaldehyde, and a wider range of substrates than those from the conventional activated sludge process (Visvanathan *et al.*, 2005).

This research studied the performance of SBR and MBR in removal of formaldehyde in synthetic wastewaters. Operational parameters that affect formaldehyde removal efficiency, i.e. contact time, formaldehyde concentration, mixed liquor suspended solids (MLSS), pH, and solid retention time (SRT), were investigated. In addition, factors affecting membrane fouling of the MBR system were studied. The results of this research can be applied to treat formaldehyde-containing wastewater in the industrial scale.

1.2 Objectives

1.2.1 To study the effects of operational variables, i.e. contact time, formaldehyde concentration, MLSS, pH, and SRT on formaldehyde removal using aerobic biological process.

1.2.2 To investigate the performance of SBR and MBR in removal of formaldehyde in synthetic wastewater.

1.2.3 To study the effect of SRT on SBR performances associated with formaldehyde removal and effluent quality.

1.2.4 To study the effect of SRT on MBR performances associated with formaldehyde removal, effluent quality, and membrane fouling.

1.3 Scopes of study

1.3.1 Wastewater used in this study was synthetic wastewater, prepared from 38-40% (w/v) formaldehyde.

1.3.2 Lab-scale SBR was set-up to simulate an activated sludge process. Sludge obtained from urea–formaldehyde resin industry was used to inoculate the system. Factor affecting the SBR performance, i.e. SRT, was studied.

1.3.3 Lab-scale MBR with submerged microfiltration (MF) membrane was set-up with the following conditions:

1) Membrane type and configuration: submerged hollow fiber–MF membrane.

2) SRT: 10, 30, and 60 days.

3) MBR performance investigation:

- Permeate quality: formaldehyde concentration, total organic carbon (TOC), and Total suspended solids (TSS).
- Specific flux: permeate volume per membrane area–time-transmembrane pressure (L/m^2-hr-KPa).

1.4 Expected results

1.4.1 To know feasibility of SBR and/or MBR in formaldehyde removal.

1.4.2 To be able to apply and upscale the SBR and/or MBR for formaldehyde removal into industrial scale.

CHAPTER 2

THEORY AND LITERATURE REVIEWS

2.1 Formaldehyde

Formaldehyde is a compound which is commonly used in the chemical industry, textile processing, paper industry and wood processing. Most formaldehyde is used in the production of polymers and other chemicals. Reaction of formaldehyde with phenol, urea, or melamine, hard thermoset resins are produced. The resins are commonly used in permanent adhesives for plywood or carpeting. They are also foamed to make insulation, or casted into molded products. Production of formaldehyde resins accounts for more than half of formaldehyde consumption. Those industries produce wastewater with high concentration of this compound for example, during the production of polyester fibers (3–5 g formaldehyde/L), industrial adhesives (0.2–40 g formaldehyde/L) (Campos *et al.*, 2002). It is frequently released in wastes causing environmental pollution (Adroer *et al.*, 1989). Formaldehyde is considered as a very toxic agent with severe negative action on living organisms. Because of its high toxicity, formaldehyde is often used as an active ingredient in preservatives and disinfectant agents to inhibit microbial activity (Eiroa *et al.*, 2005). Industrially, formaldehyde is produced by the catalytic oxidation of methanol. The most commonly catalysts are silver metal and a mixture of an ion oxide with molybdenum. In the more commonly used iron oxide system, methanol and oxygen react at 400 °C to produce formaldehyde according to the chemical equation as shown in Eq. 2.1.

$$CH_3OH + \frac{1}{2}O_2 \rightarrow HCHO + H_2O \qquad (2.1)$$

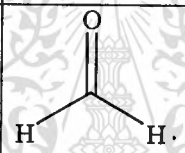
The silver–based catalyst is usually operated at a higher temperature, about 650 °C on it, two chemical reactions simultaneously produce formaldehyde, as shown in Eq. 2.2

$$CH_3OH \rightarrow HCHO + H_2 \qquad (2.2)$$

Formaldehyde reacts with water to form an equilibrium mixture of water, formaldehyde, and formaldehyde hydrate shown in Eq. 2.3. The 37% solution of formaldehyde and water is known as formalin. Physical and chemical properties of formaldehyde are listed in Table 2.1

$$\underset{\text{Formaldehyde}}{CH_2O(g)} + H_2O\,(l) \rightleftharpoons \underset{\text{formaldehyde hydrate}}{CH_2(OH)_2\,(l)} \qquad (2.3)$$

Table 2.1 Physical and chemical properties of formaldehyde

Properties	Value/Characteristic
1. Chemical name	Formaldehyde
2. Synonym	Formic aldehyde, methanal, methylaldehyde, methylene oxide
3. Chemical formula	CH_2O
4. Chemical structure	
5. Molecular weight	30.03
6. Appearance	Colorless liquid, typically 37% formaldehyde in water
7. pH	2.8–4.0 (25 degrees Celsius) (37% solution)
8. Boiling point	-19.3 °C (pure formaldehyde) 98 °C (37% formaldehyde)
9. Melting point	-117 °C
10. Vapour pressure	55 mmHg at 37 °C
11. Vapour density	1 (Air = 1)
12. Specific gravity	1.08 (water = 1)
13. Log Kow	0.350
14. Log Koc	1.567
15. Solubility in water	> 100 g/100 ml at 20 °C
16 Henry's law constant	3.27×10^{-7} atm.m^3/mol
17. Ionicity in water	Non–ionic

Source: Agency for Toxic Substances and Disease Registry (2009)

2.2 Activated sludge process

The activated sludge process is used routinely for biological treatment of municipal and industrial waste waters. It is performed by a variable and mixed community of microorganisms in an aerobic aquatic environment. These microorganisms derive energy from carbonaceous organic matter in aerated wastewater for the production of new cells in a process, known as synthesis, while simultaneously releasing energy through the conversion of this organic matter into compounds that contain lower energy, such as carbon dioxide and water, in a process called respiration. In addition, a variable number of microorganisms in the system obtain energy by converting ammonia nitrogen to nitrate nitrogen in a process called nitrification. This consortium of microorganisms, the biological component of the process, is known collectively as activated sludge (Metcalf and Eddy, 2004).

The overall goal of the activated sludge process is to remove substances that have a demand for oxygen from the system. This is accomplished by metabolic reactions (synthesis–respiration and nitrification) of the microorganisms, separation and settling of activated sludge solids to create an acceptable quality of secondary wastewater effluent, as well as collection and recycling of microorganisms back into the system or the removal of excess microorganisms from the system.

2.2.1 Conventional activated sludge

A conventional activated sludge process includes the following components as shown in Fig. 2.1 (Bitton, 1999).

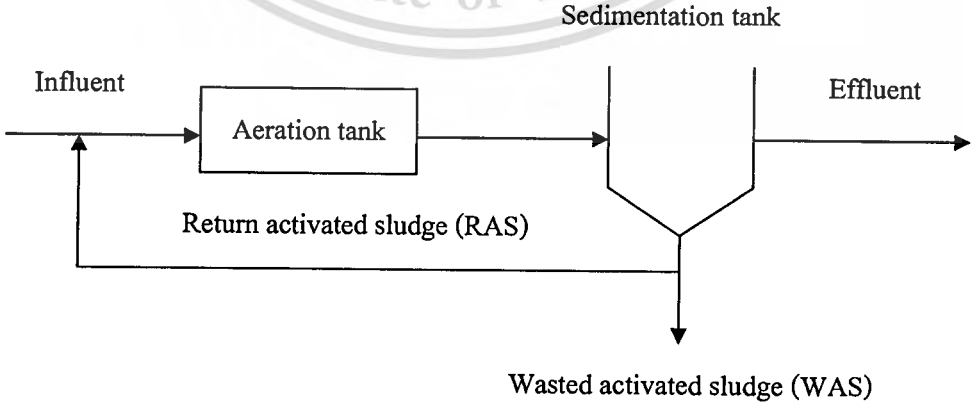


Figure 2.1 Conventional activated sludge system

1) Aeration tank: Aerobic oxidation of organic matter is carried out in this tank. Influent is introduced and mixed with return activated sludge (RAS) to form of mixed liquor, which contains 1500–2500 mg/L of suspended solids. Aeration is provided by mechanical means. An important characteristic of the activated sludge process is recycling of a large proportion of the biomass. This makes the mean cell residence time (θc) or sludge age much greater than hydraulic retention time (HRT). This practice helps maintain a large number of microorganisms that effectively oxidize organic compounds in a relatively short time. The hydraulic retention time of the aeration tank varies between 4 and 8 hours.

2) Sedimentation tank: Sedimentation tank is used for settling of microbial flocs (sludge) which are produced during the oxidation phase in the aeration tank. A portion of the sludge in the clarifier is recycled back to the aeration tank and the remainder is wasted to maintain a proper food to microorganism (F/M ratio Bitton, 1999).

2.2.2 Sequencing Batch Reactor (SBR) Process

Sequencing batch reactor (SBR) process employs a fill-and-draw reactor with complete mixing during the batch reaction step (after filling). Typically, SBR systems have four steps, which are carried out in sequence as follows:

1) Fill: During the fill operation, raw wastewater or primary effluent are added to the reactor. The fill process typically allows the liquid level in the reactor to rise from 75% of capacity to 100%. During fill, the reactor may be mixed only or mixed and aerated to promote biological reactions with the influent wastewater.

2) React: During the react period, the biomass consumes the substrate under controlled environment conditions.

3) Settle: Solids are allowed to separate from the liquid under quiescent condition resulting in a clarified supernatant that can be discharged as effluent.

4) Draw: Clarified effluent is removed during the decant period. Many types of decanting mechanisms can be used with the most popular being floating or adjustable weirs.

Each of these steps is illustrated on Fig. 2.2 (Grady *et al*, 1999).

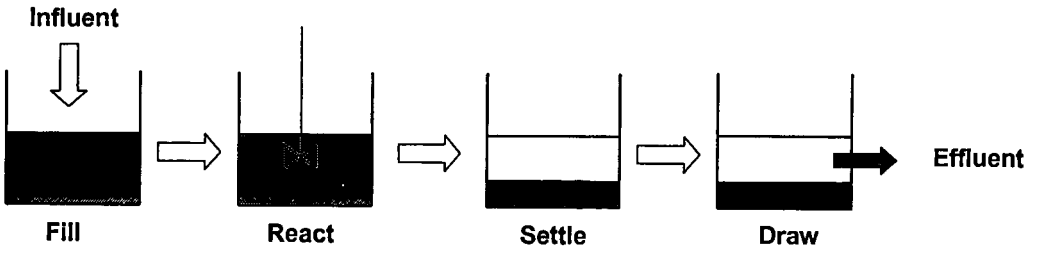


Figure 2.2 Sequencing batch reactor (SBR) process (Grady *et al.*,1999)

2.2.3 Design and operating parameters

Numerous parameters have been found to affect the performance of activated sludge processes (Cheremisinoff, 1994; Bitton, 1999). The following parameters are required to be considered in the operation of the activated process. Table 2.2 lists the values of operating parameters in various types of activated sludge process.

1) Mixed liquor suspended solids (MLSS): MLSS is the total amount of organic and mineral suspended solids, including microorganisms, in the mixed liquor. It is determined by filtering an aliquot of mixed liquor, drying at 105°C, and determining the weight of solids in the sample.

2) Mixed liquor volatile suspended solids (MLVSS): The organic portion of MLSS is represented by MLVSS, which comprises non-microbial organic matter, as well as dead and living microorganisms and cellular debris. MLVSS is determined after heating of dried filtered samples at 600–650°C, and represents approximately 65–75% of MLSS.

3) Food–to–microorganism (F/M) ratio: The F/M ratio indicates the organic load into the activated sludge system. It is expressed in kilogram BOD per kilogram of MLSS per day, as shown in Eq. 2.4.

$$\frac{F}{M} = \frac{QxBOD}{MLSSxV} \tag{2.4}$$

where Q = flow rate of sewage in million gallons par day (MGD)

BOD = 5–day biochemical oxygen demand (mg/L)

MLSS = mixed liquor suspended solids (mg/L)

V = volume of aeration tank (gallons)

For conventional aeration tanks, the F/M ratio is 0.2–0.5 kg BOD_5/kgMLSS/day but it can be higher ($\leq$1.5) for activated sludge using high–purity oxygen.

Table 2.2 Typical design parameters for commonly used activated-sludge processes

Process type	Type of reactor	SRT, d	F/M, kg BOD/kg MLSS.d	Volumetric Loading		MLSS, mg/L	Total t, h	RAS, % of influent
				lb BOD/ 1000 ft^3.d	kg BOD/ m^3.d			
High-rate aeration	Plug flow	0.5-2	1.5-2.0	75-150	1.2-2.4	200-1,000	1.5-3	100-150
Contact stabilization	Plug flow	5-10	0.2-0.6	60-75	1.0-1.3	1,000-3,000	0.5-1	50-150
						6,000-10,000	2-4	
High-purity oxygen	Plug flow	1-4	0.5-1.0	80-200	1.3-3.2	2,000-5,000	1-3	25-50
Conventional plug flow	Plug flow	3-15	0.2-0.4	20-40	0.3-0.7	1,000-3,000	4-8	25-75
Step feed	Plug flow	3-15	0.2-0.4	40-60	0.7-1.0	1,500-4,000	3-5	25-75
Complete mix	CMAS	3-15	0.2-0.6	20-100	0.3-1.6	1,500-4,000	3-5	25-100
Extended aeration	Plug flow	20-40	0.04-0.10	5-15	0.1-0.3	2,000-5,000	20-30	50-150
Oxidation ditch	Plug flow	15-30	0.04-0.10	5-15	0.1-0.3	3,000-5,000	15-30	75-150
Batch decant	Batch	12-25	0.04-0.10	5-15	0.1-0.3	2,000-5,000	20-40	NA
Sequencing batch reactor	Batch	10-30	0.04-0.10	5-15	0.1-0.3	2,000-5,000	15-40	NA
Countercurrent aeration system (CCAS™)	Plug flow	10-30	0.04-0.10	5-10	0.1-0.3	2,000-4,000	15-40	25-75

Source: Metcalf and Eddy (2004)

4) Hydraulic retention time (HRT): The hydraulic retention time is an average time spent by the influent liquid in the aeration tank of the activated sludge process. It is expressed as shown in Eq. 2.5.

$$HRT = \frac{1}{D} = \frac{V}{Q} \quad (2.5)$$

where V = volume of the aeration tank

Q = flow rate the influent wastewater

D = dilution rate

5) Sludge age (θc) or Solid retention time (SRT) : Sludge age is the mean residence time of microorganisms the system. While the hydraulic retention time may be on the order of hours, the mean cell residence time may be in the order of days. Sludge age is given by Eq. 2.6.

$$\text{Sludge age (day)} = \frac{MLSS \times V}{SS_e \times Q_e \times SS_w \times Q_w} \quad (2.6)$$

where MLSS = mixed liquor suspended solids (mg/L)

V = volume of aeration tank (L)

SSe = suspended solids in wastewater effluent (mg/L)

Qe = quantity of wastewater effluent (m^3/day)

SSw = suspended solids in wasted sludge (mg/L)

Qw = quantity of wasted sludge (m^3/day)

Sludge age may vary from 5 to 15 days in conventional activated sludge. It varies with the season of the year and is higher in the winter than in the summer season.

6) Biochemical oxygen demand (BOD): BOD can be used as a measure of incoming food getting into aeration tank, and describe the aeration tank in terms of BOD/MLVSS/day, which is sometimes called the F/M.

2.2.4 Factors affecting organic matter oxidation in activated sludge process. (Grady *et al*, 1999)

1) Nutrients: Biological activity of sludge flocs and their setting characteristics are affected by wastewater composition. In conventional activated sludge, a COD:N:P ration of 200:5:1 is required to maintain the optimal nutrient balance for heterotrophic activity which is equivalent to an operational range of 0.03–0.06 kg nitrogen and 0.007–0.01 kg potassium per kg BOD.

2) Mixing: Mixing is required to bring organisms, oxygen, and nutrients together and to remove metabolic waste products. If there is not enough mixing, proper treatment will not take place because of lack of contact between the microorganism, their food and oxygen. If too much mixing is provided, it can cause break up of floc or formation of unstable floc particles.

3) pH: The enzymes which regulate many of the biochemical reaction in bacteria are very pH dependent. The optimum pH should be between 7.0 and 7.5 for the proper activated sludge microorganisms to dominate.

4) Temperature: The temperature needs to be maintained between 25°C and 35°C. If temperature exceeds 40°C, then the dominant microbes will be thermophilic which are thought to be less efficient than non–thermophilic microbes in this type of treatment.

5) Toxicity: Microorganisms are sensitive to a wide range of organic and inorganic compounds. In many cases, nitrification rates are inhibited even though bacteria continue to grow and oxidize ammonia and nitrite, but at significantly reduced rates. In some cases, toxicity may be sufficient to kill the nitrifying bacteria. Toxic and inhibitory inorganic and organic compounds of concerns for microorganisms in biological processes are listed in Table 2.3 and 2.4, respectively.

Table 2.3 Toxic and inhibitory inorganic compounds of concerns for biological processes

Substance	Moderately inhibitory concentration, mg/L	Strongly inhibitory concentration, mg/L
Sodium, Na^+	3,500-5,500	8,000
Potassium, K^+	2,500-4,500	12,000
Calcium, Ca^{2+}	2,500-4,500	8,000
Magnesium, Mg^{2+}	1,000-1,500	3,000
Ammonia-nitrogen, NH^+_4	1,500-3,000	3,000
Sulfide, S^{2-}	200	200
Copper, Cu^{2+}	N/A	0.5 (soluble) 50-70 (total)
Chromium, Cr(VI)	N/A	3 (soluble) 200-250 (total)
Chromium, Cr(III)	N/A	180-420 (total) 2 (soluble)
Nickel, Ni^{2+}	N/A	30 (total)
Zinc, Zn^{2+}	N/A	1 (soluble)

Source: Metcalf and Eddy (2004)

Table 2.4 Toxic and inhibitory of organic compound of concerns for biological processes

Compound	Concentration resulting 50 percent reduction in activity, mM
1-Chloropropene	0.1
Nitrobenzene	0.1
Acrolein	0.2
1-Chloropropane	1.9
Formaldehyde	2.4
Lauric acid	2.6
Ethyl benzene	3.2
Acrylonitrile	4.0
3-Chlorol-1,2-propanediol	6.0
Crotonaldehyde	6.5
2-Cholropropionic acid	8.0
Vinyl acetate	8.0
Acetaldehyde	10.0
Ethyl acetate	11.0
Acrylic acid	12.0
Catechol	24.0
Phenol	26.0
Aniline	26.0
Resorcinol	29.0
Propanol	90.0

Source: Metcalf and Eddy (2004)

2.2.5 Biological component of the activated sludge system

The biological component of the activated sludge system is comprised of microorganisms. The composition of these microorganisms is 70 to 90 percents organic matter and 10 to 30 percents in organic matter. Cell makeup depends on both the chemical composition of the wastewater and the specific characteristics of the organisms in the biological community. Bacteria, fungi, protozoa, and rotifers constitute the biological component, or biological mass, of

activated sludge. In addition, some *metazoa*, such as nematode worms, may be present. However, the constant agitation in the aeration tanks and sludge recirculation are deterrents to the growth of higher organisms. The species of microorganism that dominates a system depends on environmental conditions, process design, the mode of plant operation, and the characteristics of the secondary influent wastewater. The microorganisms that are of greatest numerical importance in activated sludge are bacteria, which occur as microscopic individuals from one micron in size to visible aggregations or colonies of individuals. Some bacteria are strict aerobes (they can only live in the presence of oxygen), whereas others are anaerobes (they are active only in the absence of oxygen). The predominance of bacteria living in activated sludge are facultative-able to live in either the presence or absence of oxygen, an important factor in the survival of activated sludge when dissolved oxygen concentrations are low or perhaps approaching depletion. While both heterotrophic and autotrophic bacteria reside in activated sludge, the former predominate. Heterotrophic bacteria obtain energy from carbonaceous organic matter in influent wastewater for the synthesis of new cells. At the same time, they release energy via the conversion of organic matter into compounds such as carbon dioxide and water. Important genera of heterotrophic bacteria include *Achromobacter, Alcaligenes, Arthrobacter, Citromonas, Flavobacterium, Pseudomonas, and Zoogloea.* The bacteria form two types of sludge depending on how the bacteria form communities. There are bacteria that form flocs or conglomerates that sink to the bottom forming a dense sludge and a clear effluent. There are also filamentous bacteria that develop with fungi. The most dominant type of bacteria are filamentous *Haliscomenobacter hydrossis* that is an indicator of low oxygen content. Another less common type of bacteria is the *Thiothrix spp.*, which is a sulphur based genus. The bacteria are prey to larger microorganisms, including amoeba *(Sarcodira)*, free swimming ciliates *(ciliata),* stalked ciliates *(vorticela),* and *suctoria.* There are also multicellular rotifer organisms in the activated sludge. Fig. 2.3 illustrates the relative abundance and development of microorganisms communities in a typical activated sludge aeration tank (Grady *et al*, 1999).

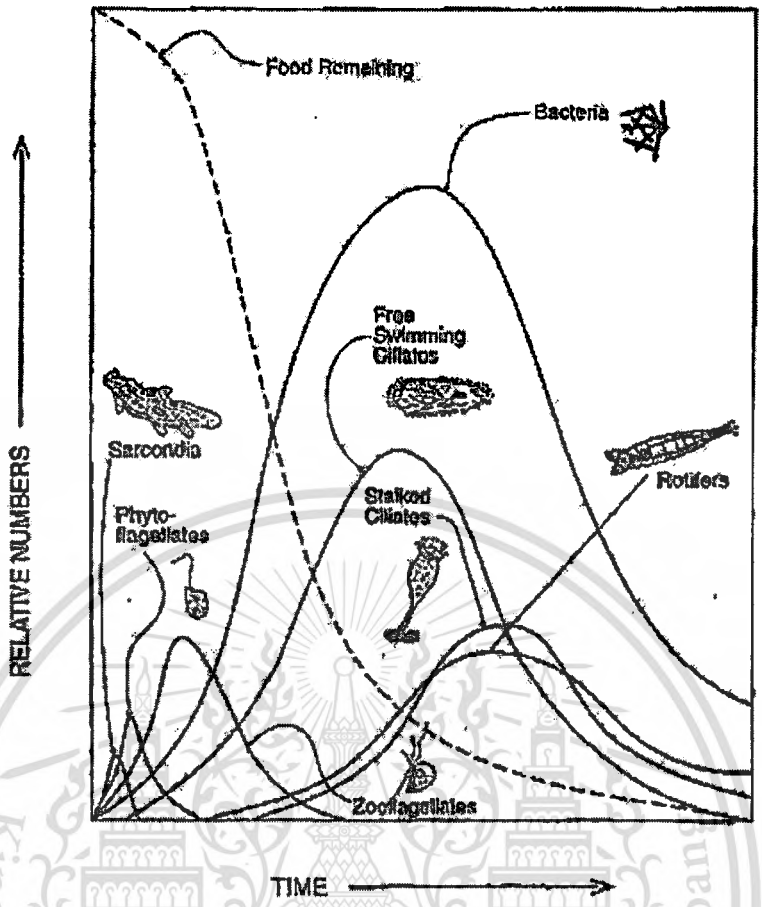
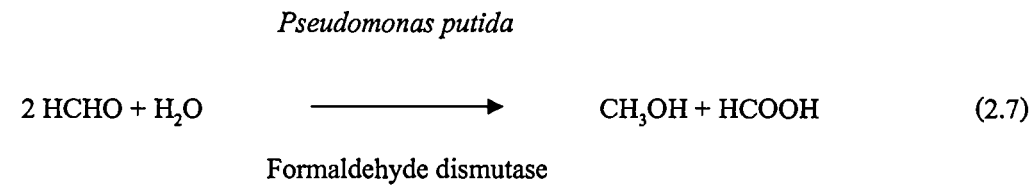
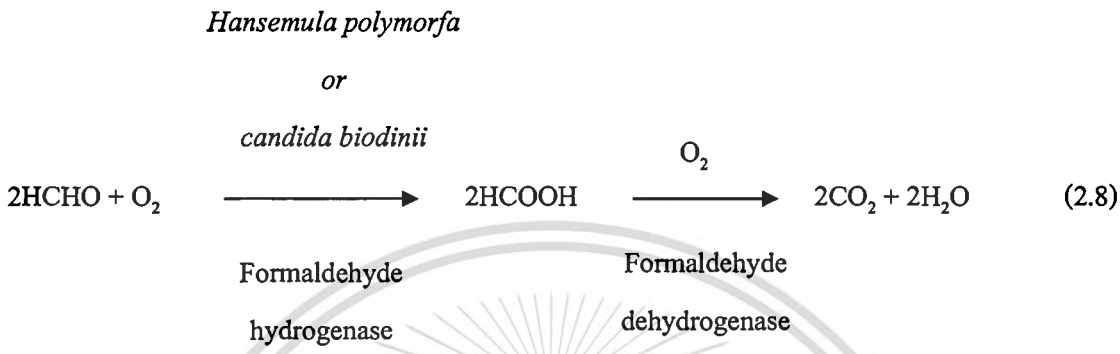


Figure 2.3 Predominance of microorganisms in the aeration tank (http://www.geocities.com/RainForest/Vines/4301/bugs.html200713)

Formaldehyde is considered to be a very toxic substance to most microorganisms. However, some strains of microorganisms can resist and survive in the media containing formaldehyde (Glancer–Soljan et al., 2001). Aerobic biodegradation of formaldehyde is achieved by specific strains of bacteria, for example *pseudomonas spp.* Formic acid is the basic intermediate in formaldehyde biodegradation. *Pseudomonas spp.* degrades formaldehyde with formaldehyde dismutase enzyme, while yeast, e.g. *Hansenula spp.* and *Candida spp.* genera, achieves it using the enzymes formaldehyde and formate dehydrogenase. *Trichosporon penicilatum* in mixed culture plays a significant role in forming biofloc, leading to settling of biosludge (Glancer-Soljan *et al.*, 2001). The transformations of formaldehyde are shown in Eqs. 2.7 and 2.8:



(2.7)



(2.8)

2.3 Membrane process

A membrane or, more properly, a semipermeable membrane, is a thin layer of material that is capable of separating materials as a function of their physical and chemical properties when driving forces employed. For example, microfiltration (MF) and reverse osmosis (RO) are two membrane processes that use pressure to transport water across the membrane. MF membranes are capable of removing only particulate matter, while RO membranes retain many solutes as water permeates through the membrane. Electrodialysis is also capable of separating ionic solutes from water, but, in this case, ions are transported across the membrane and driving force is an electrical potential (Mallevialle *et al.*, 1996).

2.3.1 Membrane operation

In membrane processes there are three possible streams: a feed, a retentate and a permeate stream. The retentate stream is defined as unpermeated product. If there is no retentate stream, the operation is termed dead–end or full–flow, as shown in Fig. 2.4(a). Such operation is normally restricted to either low–solids water, for example, cartridge filtration of boiler feed water or ultrafiltration for pure water production, or cyclic operation with frequent backwashing. For waters having a significant solids loading and membranes of limited permeability (dense membranes), it is not desirable to try and convert all of the feed to permeate product in a single passage through a module. In such cases, cross–flow operation is employed, as shown in Fig. 2.4(b), where some of the feedwater is collected as a concentrate (or retentate) stream.

This expedites the removal of accumulated materials from the membrane–solution interfacial region provided by the scouring action of the retentate flowing over the membrane surface (Simon and Bruce, 2003).

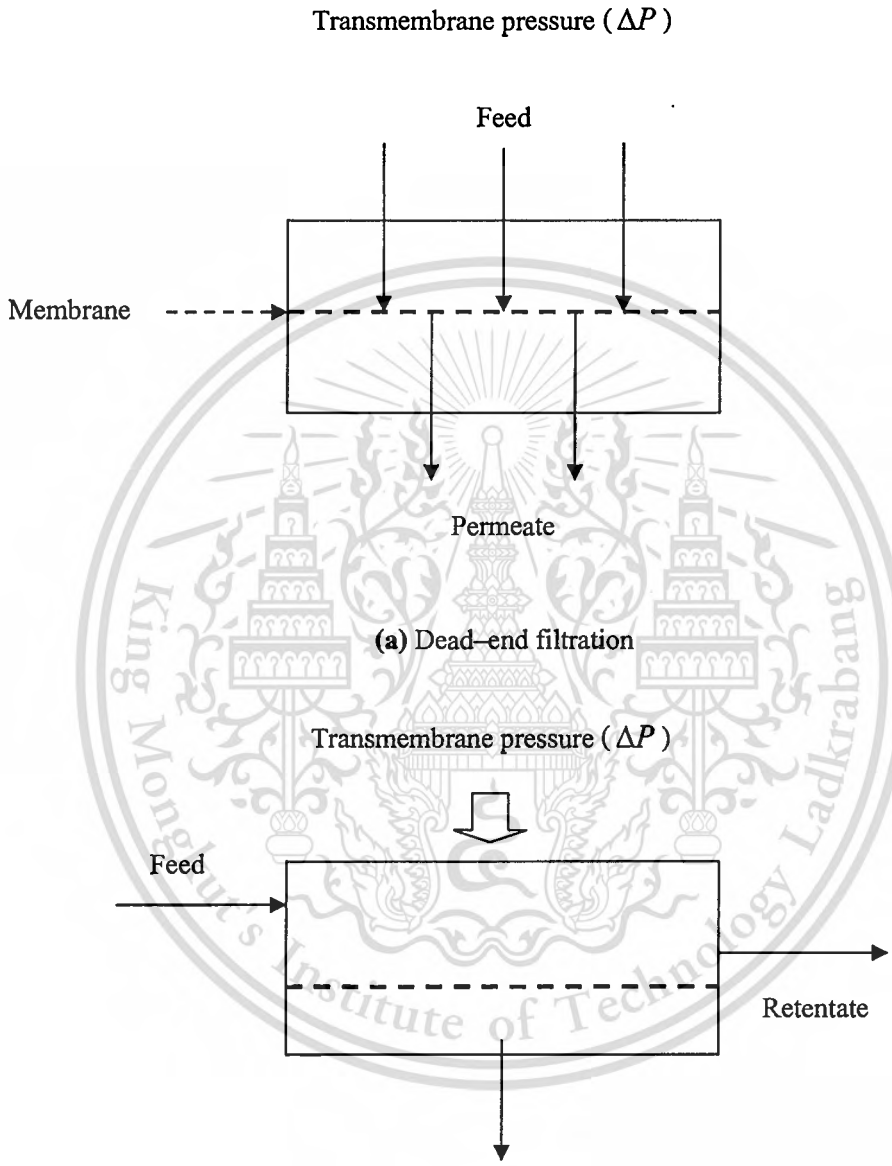


Figure 2.4 Modes of membrane operation

2.3.2 Membrane fouling

The major symptoms of membrane fouling are flux decline over operating time, increasing transmembrane pressure, sludge cake formation, and changes to the retention coefficient (Wiesmann *et al.*, 2007). A reduction in membrane flux density is caused by accumulation of foreign materials. Such materials may accumulate on, or in, the neighborhood of the membrane surface, or it becomes embedded within the membrane pores, or it may cause changes in the chemical character of the membrane material. Almost all substances in water have the potential to foul membranes. These substances may be categorized as:

1) particles.

2) organic compounds.

3) inorganic compounds.

4) biota that grow on the membrane surface.

Particles foul membranes by either accumulating inside membrane pores or by blocking pores due to surface deposition. Organics, inorganics, and biota may adsorb to membrane surfaces and pores. The nature and extent of fouling is influenced by the chemical and biological characters of solution chemical composition of the membrane, solute–solute type interactions, and membrane–solute type interactions (Hendricks, 2005).

In case of MBR, different fouling mechanisms are shown in Fig. 2.5. An irreversible sludge cake layer is formed by particles, contaminants, and agglomerates of contaminants which are bigger than the pore size of the membrane. Due to the heterogeneous nature of bioreactor, mixed liquor fouling is difficult to predict and control in a MBR. Factors affecting membrane fouling in the MBR are including (Wisemann *et al.*, 2007):

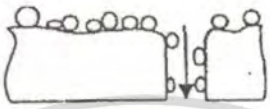
1) The membrane material hydrophobicity, porosity, and pore size and distribution.

2) The mass of microorganism (MLSS) and of extra cellular polymeric substances (EPS), floc structure, dissolved matter, and floc size.

3) The operation conditions, e.g. configuration, cross flow velocity, aeration, hydraulic and solid retention time, and trans–membrane pressure.

Pore size of the membrane small than substances

Pore size of the membrane bigger than substances



Pore size of the membrane bigger than substances

Figure 2.5 Membrane fouling mechanisms (Martinez-Diez *et al.*, 1996).

2.4 Membrane bioreactor

Membrane bioreactor (MBR) is a combination of suspended growth activated sludge biological treatment and membrane filtration equipment performing the critical solids/liquid separation function. Traditional activated sludge system uses an aeration tank and secondary clarifier. In MBR, the secondary clarifiers are replaced by microfiltration (MF), ultrafiltration (UF) membranes (Water Environment Federation, 1994). There are two general types of membrane systems that can be used in MBR: pressure–driven, in–pipe cartridge systems that are located external to the bioreactor and vacuum–driven, immersed systems that are designed for installation within the bioreactor, as shown in Fig. 2.6 and 2.7, respectively. Immersed membrane technologies using hollow–fiber or flat–sheet membranes are the most popular for MBR applications because they operate at lower operating pressures (or vacuums), can more readily accommodate the variations in the types of solids found in activated sludge bioreactors, and typically provide a lower life–cycle cost, particularly for municipal-scale facilities. Pressure–driven systems are more prevalent in industrial system where wastewater characteristics, such as high temperatures, require the use of ceramic membranes. In its simplest form, an immersed membrane bioreactor system can combine the functions of an activated sludge aeration system, secondary clarifiers, and filtration in a single tank.

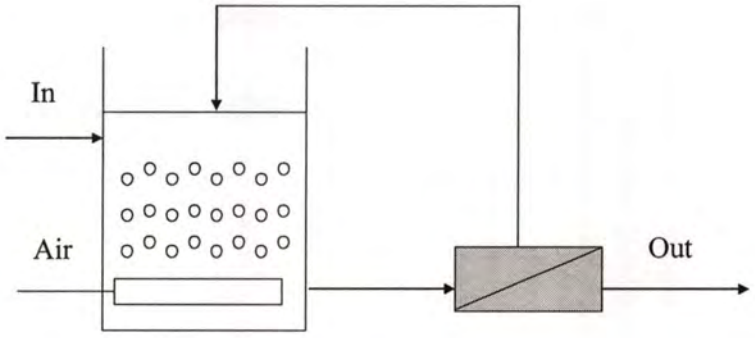


Figure 2.6 External MBR schematic (Water Environment Federation, 1994).

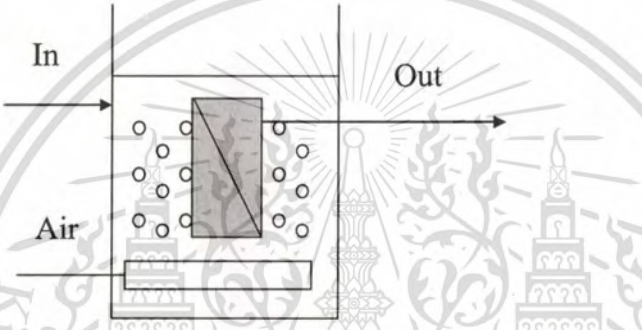


Figure 2.7 Immersed MBR schematic (Water Environment Federation, 1994).

Immersed membrane technologies are found to be popular for membrane bioreactor (MBR). Two major types of membrane modules are plate and frame, and hollow fiber.

2.4.1 Plate and frame

These modules are made up of stacked flat–sheet membranes and support plates, as shown in Fig. 2.8. The feed circulates between the membranes of two adjacent plates. The thickness of the liquid sheet is in the range of 0.5 to 3.0 mm. The packing density of plate–and–frame units is about 100 to 400 m^2/m^3. The plates ensure the mechanical support of the membrane and, at the same time, the drainage of the permeate. The plates may be corrugated on the feed side to improved mass transfer. Their arrangement makes it possible to bring about, in parallel and/or in series, circulation. Large unitary assemblies with a surface of up to 100 m^2 can be formed. The units are easily disassembled to gain access for manual cleaning or replacement to the membrane (Mallevialle *et al.*, 1996).

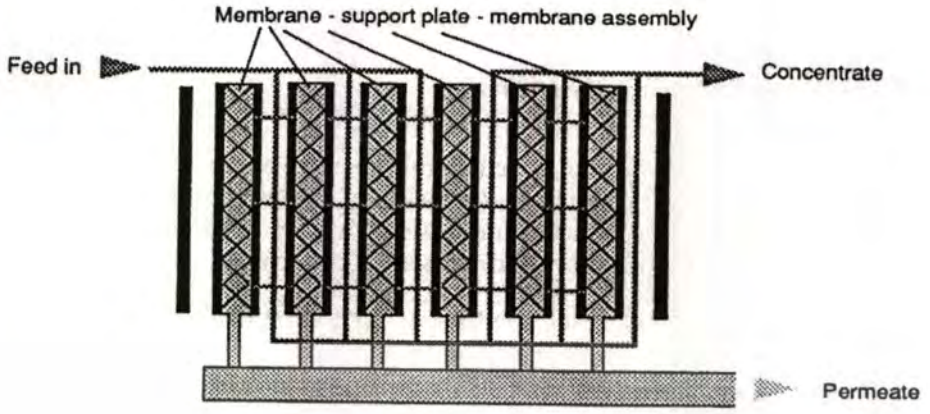


Figure 2.8 Plate and frame membranes (Mallevialle *et al.*, 1996)

2.4.2 Hollow fiber

The fibers are gathered in a bundle of several thousand, even several million. Flow of the feed tanks place either inside the fibers (inside–out configuration) or out side the fibers (outside–in configuration). In the first case, the water tightness between the feed and permeate flows is provided by a potting resin which forms a tube plate at each of the bundle. After hardening of the resin, the bundle is cut in such a way that the open ends of all the fibers appear, as shown in Fig. 2.9. In the outside–in configuration, the bundle is often arranged in a U-shape, and the fibers are sealed at only one end, as shown in Fig. 2.10 (Mallevialle *et al.*, 1996).

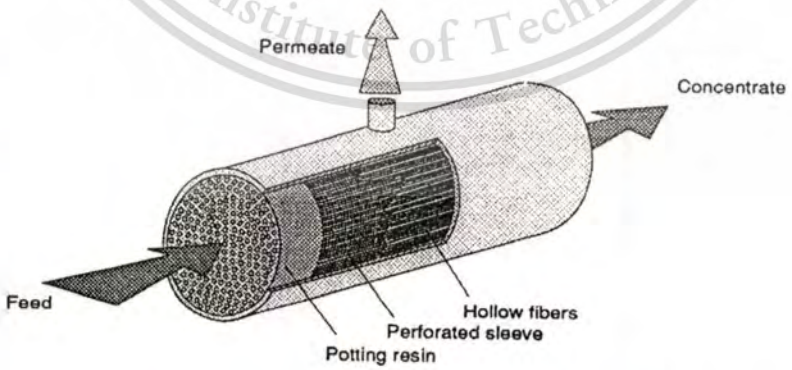


Figure 2.9 Hollow fiber membrane (Mallevialle *et al.*, 1996).

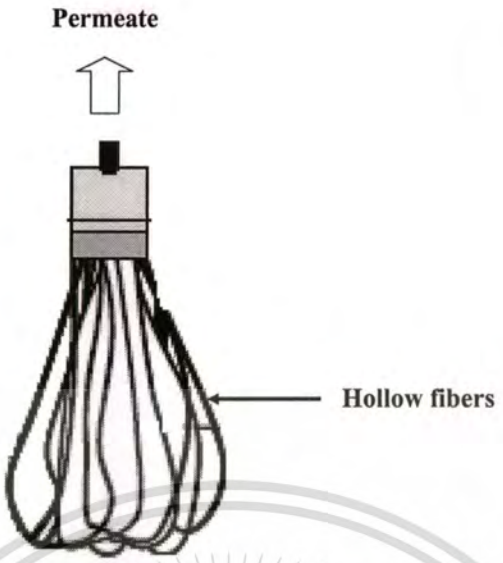


Figure 2.10 U-shape outside-in hollow fiber membrane (Adapted from Lim *et al.*, 2007)

Comparison between plate and frame and hollow fiber membrane are listed in Table 2.5.

Table 2.5 Comparison between plate and frame and hollow fiber membrane

Parameter	Plate and frame membrane	Hollow fiber membrane
Cost	Moderate	Low
Specific surface area (m^2/m^3)	200-500	500-30,000
Energy consumption	Moderate	Low
Fouling tendency	Low	High
Fouling control	Moderate	Moderate to Good
Cleaning	Moderate	Difficult
Membrane replacement	Easy	Impossible

Source: Tung (2007)

2.5 Literature reviews

Adroer *et al.* (1989) studied formaldehyde biodegradation by a strain of *Pseudomonas putida.* The results indicated that the biodegradation was initiated by a dismutation reaction, yielding as products formic acid and methanol. The degradation of methanol and formic acid began after exhaustion of formaldehyde in the medium, and presened a diauxic pattern: first

formic acid was consumed, followed by methanol. Moreover, cell viability, which was affected by the amount of added formaldehyde, had been determined.

Omil *et al.* (1999) studied degradation of formaldehyde in anaerobic batch digesters in the presence and the absence of a co-substrate composed of volatile fatty acids. No adapted sludge were used with specific maximum methanogenic activities on volatile fatty acids around 0.6 g chemical oxygen demand (COD)g^{-1} volatile suspended solids (VSS) day^{-1} and with a significant sulfate-reducing activity which led to the consumption of around 12% COD when sulfate was present. Formaldehyde degradation was favored by the presence of the co-substrate especially when high acetate concentrations were used (30 mM vs. 10 mM). Both in the absence and the presence of the co-substrate, methanol was shown to be a key intermediate. Formaldehyde exerts a clear inhibitory effect on volatile fatty acids degradation. Values of 4.17 mM (125 mg formaldehyde/L) exert a 50% decrease in the specific sludge activity. When formaldehyde concentrations ranged from 5–6.67 mM (150–200 mg formaldehyde/L), methanol was accumulated in the medium.

Glancer-soljan *et al.* (2001) used selected strains of *Pseudomonas putida, Pseudomonas cepacia*, *Trichosporon penicillatum* yeast and the mixed culture of these three strains for aerobic degradation of formaldehyde and formic acid in the synthetic medium and in wastewater generated by melamine resins production. It was found that the mixed culture in the synthetic medium degraded 1000 mg/L of formaldehyde over 18–24 hours and 500 mg/L of formic acid over 12–18 hours. Aerobic degradation of wastewater from the production of melamine resins with the use of mixed bacterial and yeast culture was achieved in 24 hours with COD reduction of over 90 % and complete degradation of formaldehyde, methanol and butanol. The role of *Trichosporon penicillatum* yeast in the mixed culture, during aerobic degradation of formaldehyde in the synthetic medium and wastewater, was to form flocculent biomass, leading to self-precipitating.

Hong *et al.* (2002) studied factors affecting membrane fouling during membrane bioreactors (MBR) operation. It was found that larger pore size membranes (MF) experienced higher initial fouling, presumably due to pore blocking. Fouling was independent of MLSS concentration for the range between 3600-8400 mg/L. Initial fouling rates were higher for high suction TMP, indicating that initial permeation rate was very important in controlling membrane fouling in MBR processes. Intermittent suction operation resulted in slower flux declines due to enhanced removal of foulants accumulated on the membrane surface.

Eiroa *et al.* (2004) studied formaldehyde biodegradation, urea hydrolysis and denitrification in anoxic batch assays and in a continuous laboratory anoxic reactor were investigated. In batch assays, the initial formaldehyde biodegradation rate was around 0.7 g CH_2O g^{-1}VSS d^{-1} and independent of the urea concentration (90–370 mg N–NH_2CONH_2/L). Urea was completely hydrolyzed to ammonium in the presence of 430 mg/L formaldehyde and complete denitrification took place in all cases (125 mg N–NO_3^-/L). Formaldehyde removal efficiencies above 99.5% were obtained in a lab-scale denitrifying up-flow sludge blanket reactor at organic loading rates between 0.37 and 2.96 kg COD m^{-3} d^{-1} (625–5000 mg CH_2O/L). The urea loading rate was increased from 0.06 to 0.44 kg Nm^{-3} d^{-1} (100–800 mg N–NH_2CONH_2/L) and hydrolysis to ammonium was around 77.5% at all loading rates. The denitrification process was always almost complete (100–800 mg N–NO_3^-/L), due to the high COD/N ratio of 6.7 in the influent. A minimum value of 3.5 was found to be required for full denitrification. The composition of the biogas indicated that denitrification and methanogenesis occurred simultaneously in the same unit. A good granulation of the sludge was observed.

Marin *et al.* (2008) investigated causes of membrane fouling in membrane bioreactors (MBR). The results showed that membrane fouling was caused by cake formation on the surface, mainly attributed to suspended solids, and also by surface adsorption connected with pore blocking, attributed to soluble components of activated sludge. The irreversible blocking by adsorption had been identified as the major cause of fouling, as it had gradually decreased membrane permeability from 417 to 55 L m^{-2} h^{-1} bar^{-1} in 123 days of continuous experiment without chemical cleaning. Cake resistance was small due to a low flux and high aeration intensity and it remained constant for most of the experiment. A sudden acceleration of fouling had been observed after 119 days of operation and attributed to irreversible fouling that caused local fluxes to exceed critical flux and led the filtration into critical conditions, where suspended solids started to deposit. Faster fouling, in comparison to the filtration of normal sludge, had been observed when starving biomass was filtered. That was attributed to floc rupture.

CHAPTER 3

RESEARCH METHODOLOGY

3.1 Chemicals

1. Sodium hydroxide (NaOH), analytical grade from Lab Scan, Thailand.
2. Sulfuric acid (H_2SO_4), analytical grade from Carlo Erba, Italy.
3. Ammonium acetate (CH_3COONH_4), analytical grade from Carlo Erba, Italy.
4. Acetic acid glacial (CH_3COOH), analytical grade from Lab Scan, Thailand.
5. Acetylacetone ($C_5H_8O_2$), analytical grade from Fluka Chemika, Switzerland.
6. Thymolphthalein ($C_{28}H_{30}O_4$), analytical grade from Fluka Chemika, Switzerland.
7. Sodium sulphite anhydrous (Na_2SO_3), analytical grade from Ajax Finechem, Austria.
8. Formaldehyde solution (HCHO), analytical grade from Ajax Finechem, Austria.
9. Ethanol 95% (C_2H_5OH), analytical grade from Lab Scan, Thailand.
10. Calcium chloride anhydrous ($CaCl_2.2H_2O$), analytical grade from Lab Scan, Thailand.
11. Ammonium sulphate ($(NH_4)_2SO_4$), analytical grade from Carlo Erba, Italy.
12. Magnesium sulphate ($MgSO_4.7H_2O$), analytical grade from Carlo Erba, Italy.
13. Potassium dihydrogen phosphate (KH_2PO_4), analytical grade from Carlo Erba, Italy.
14. Disodium hydrogen phosphate (Na_2HPO_4), analytical grade from Carlo Erba, Italy.

3.2 Apparatus and Instruments

1. UV – visible spectrophotometer; Heiosά, from Thermo electron, England.
2. Total organic carbon analyzer; TOC –V_{CSH}, from Shimadzu, Japan.
3. Electrical balance 4 figures; B-3100S, from Sartorius, Thailand.
4. pH meter; 215, from Denver instrument, England.
5. DO meter; 9200, from Jenway, England.
6. Air pump; ACO-208, from Hailea, China.
7. Water bath; WB 22, from Memmert, England
8. Filter paper; Whatman 0.45 μm
9. Glasswares
10. Sequencing batch reactor (SBR) system (see section 3.3.3)
11. Membrane bioreactor (MBR) system (see section 3.3.4)

3.3 Experimental Methods and Analyses

Experiments were conducted in laboratory of Chemistry Department, Faculty of Science, King Mongkut's Institute of Technology Ladkrabang. Overall scope the experiments are shown in Fig 3.1. The experiments were divided into 3 steps, including 1) Batch tests 2) SBR operation, and 3) MBR operation.

3.3.1 Source of wastewater

Wastewater used in the experiment was formaldehyde-containing synthetic wastewater. The synthetic wastewater was prepared from 38-40% (w/v) formaldehyde stock solution. The influent concentration of formaldehyde was varied from 100 to 1,000 mg/L by diluting with DI water. Formaldehyde was used as a sole carbon source. Required nutrients were also added in the synthetic wastewater for microorganism growth and metabolism. The ratio of COD:N:P are controlled at 200:5:1 (Eiroa *et al.*, 2004). The detail of preparation and composition of synthetic wastwater used in the experiment, are shown in Appendix A-1 (Table A-1, A-2, and A-3).

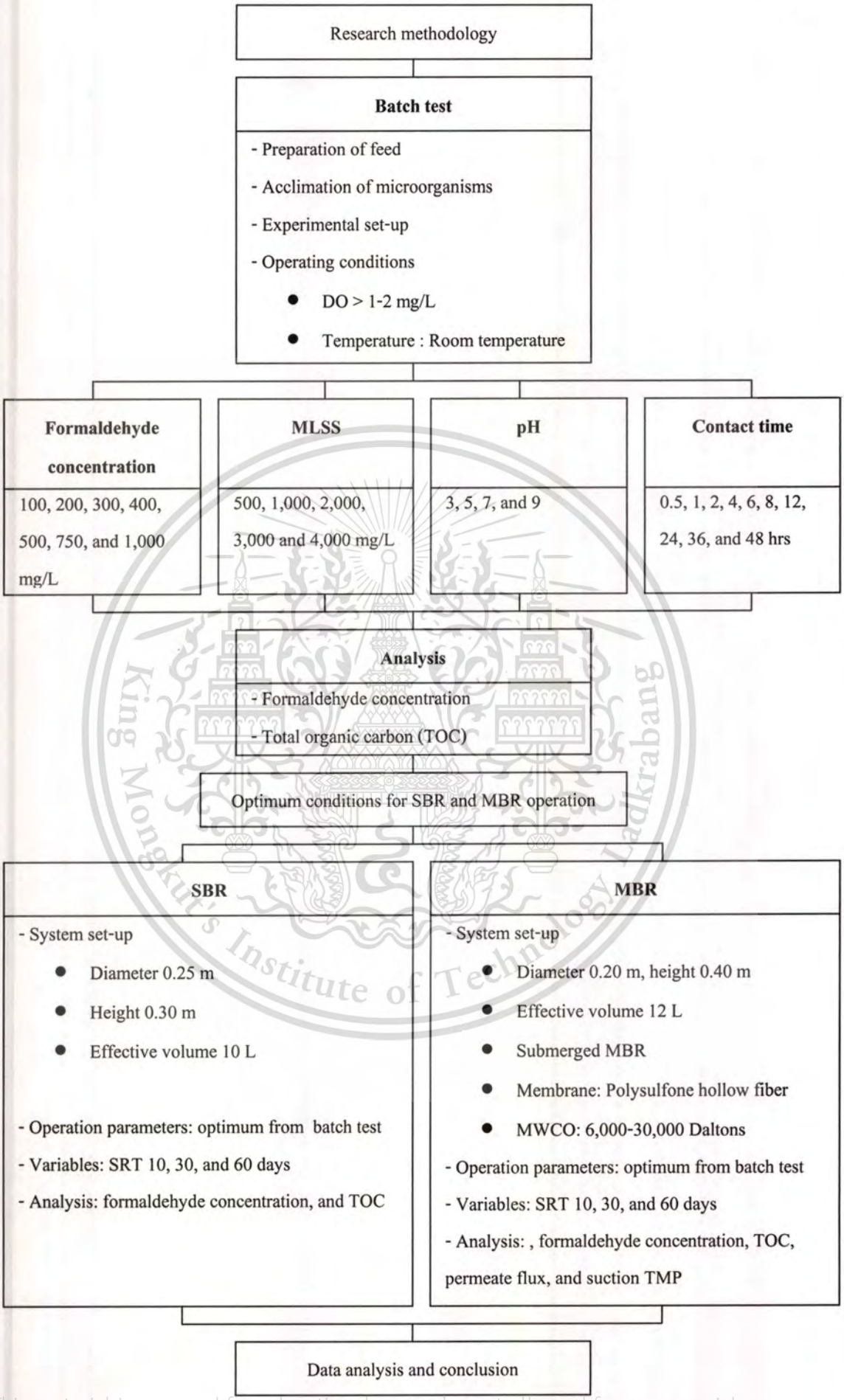


Figure 3.1 Steps of the research

3.3.2 Batch tests

1) Experimental set-up

Batch tests were conducted in reactors, made of 2.5-L amber glass bottles with an effective volume of 2 L. Air was continuously supplied from an air-pump through air diffusers, as shown in Fig 3.2. Seed sludge, obtained from wastewater treatment plant of urea-formaldehyde resin industry, was acclimated in synthetic wastewater before the experiments.

To acclimate the sludge, formaldehyde-contaminated synthetic wastewater with required nutrients (i.e. nitrogen and phosphate) was fed as substrate. The concentration of formaldehyde was increased up to approximately 500 mg/L for sludge growth. The MLSS was measured to indicate a steady state of the system.

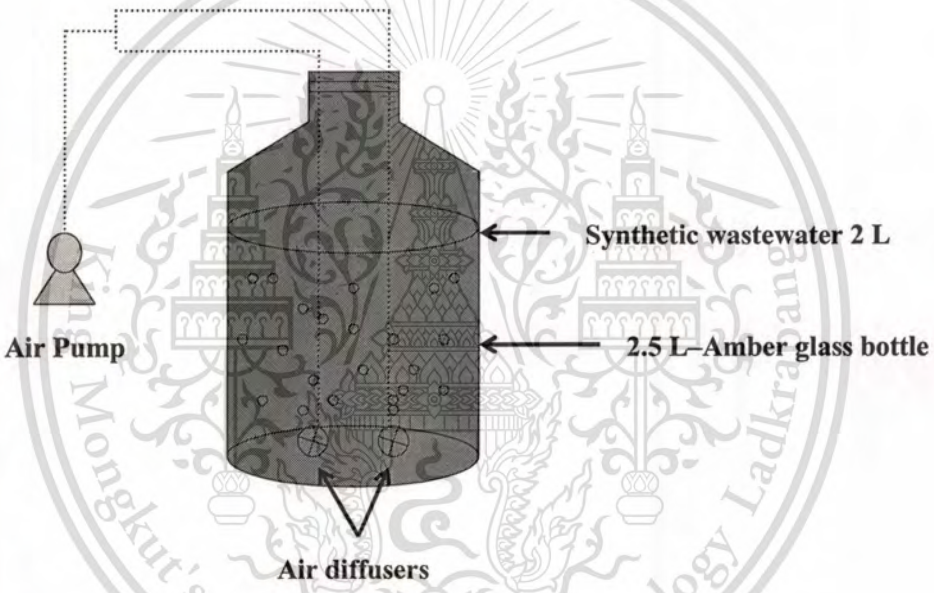


Figure 3.2 Schematic representation of the batch test experimental set-up

2) Operating conditions and variable parameters

All batch experiments were performed at room temperature (~30°C). The pH in the reactor was maintained at 7.0-7.5, unless specified. Dissolved oxygen was maintained more than 2 mg/L at all times. The effects of contact time, formaldehyde concentration, MLSS, and pH on formaldehyde removal efficiency were investigated. The variable parameters for batch tests were listed in Table 3.1. The experimental procedures are described as follows:

Table 3.1 Variable parameters for batch test experiment

Variable parameters	Unit	Values
Formaldehyde concentration	mg/L	100, 200, 300, 400, 500, 750, and 1,000
MLSS	mg/L	500, 1,000, 2,000, and 4,000
pH	-	3, 5, 7, and 9
Contact time	hrs	0.5, 1, 2, 4, 6, 8, 12, 24, 36, and 48

2.1) Effects of contact time and formaldehyde concentration

Using experiment in terms of batch test, adding 2 L of synthetic wastewater containing formaldehyde at concentrations of 100, 200, 300, 400, 500, 750, and 1,000 mg/L, into 2.5-L amber glass bottles. The acclimated sludge in each bottle was maintained with MLSS of 500 mg/L, approximately. The pH was controlled at 7 using phosphate buffer while DO was maintained more than 2 mg/L. Parallel set of experiment without sludge was conducted as a control group. Samples were taken at contact times of 6, 8, 12, 24, 36, and 48 hrs, and then filtered with 0.45-μm filter papers, prior to analyses for formaldehyde concentration and total organic carbon (TOC).

2.2) Effects of MLSS

Using experiment in terms of batch test, adding 2 L of synthetic wastewater containing formaldehyde at concentration obtained from 2.1) into 2.5-L amber glass bottles. The acclimated sludge was then added to each bottle to yield different MLSS of 500, 1,000, 2,000, and 4,000 mg/L. The pH was controlled at 7 using phosphate buffer while DO was maintained more than 2 mg/L. Control experiment was done at the same condition, but without sludge. Samples were taken at contact times of 0.5, 1, 2, 4, 6, 8, and 24 hrs, and then filtered with 0.45−μm filter paper, prior to analyses for formaldehyde concentration and TOC.

2.3) Effects of pH

Using experiment in terms of batch test, adding 2 L of synthetic wastewater containing formaldehyde at concentration obtained from 2.1) and MLSS concentration obtained from 2.2) into 2.5-L amber glass bottles. The pH was varied at 3, 5, 7, and 9. DO was maintained more than 2 mg/L. Parallel set of experiment without sludge was conducted as a control group. Samples were taken at contact times of 2, 4, 6, 8, and 24 hrs, and then filtered with 0.45−μm filter paper, prior to analyses for formaldehyde concentration and TOC.

3.3.3 Sequencing batch reactor (SBR)

1) Experimental set-up

SBR system consisted of bioreactor tank made of acrylic with a diameter of 0.25 m, 0.30 m high, and effective volume of 10 L, as illustrated in Fig 3.3. Equipments used in SBR operation are summarized in Table 3.2. SBR system was operated as a cycle with 8 hrs/cycle and 3 cycles/day. The sequence of each cycle during SBR operation was shown in Table 3.3. Acclimated sludge was introduced into the bioreactor with an initial MLSS concentration of 2,000-3,000 mg/L. Synthetic wastewater, as described in 3.3.1, was used as feed of the system. Influent from feed tank was delivered to the reactor using a pump, controlled by automatic timer. Air pump generating air bubbles through diffusers located at the bottom of the tank. The content in the SBR was mixed thoroughly by an agitator together with the air bubbles. After a specific period of aeration, air pump was automatically shut down. Then, sludge was allowed to settle and clarified effluent was drawn out from the tank by effluent pump before the next cycle started.

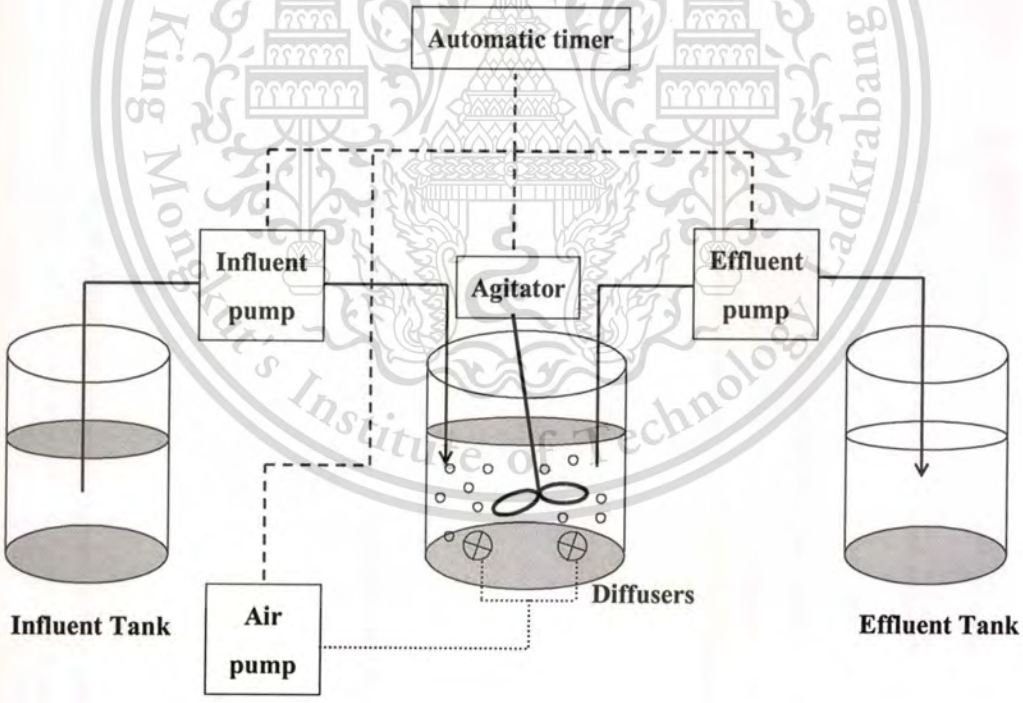


Figure 3.3 Schematic diagram of the SBR

Table 3.2 Equipments for SBR set-up

Equipment	Detail
Feed tank	80 L polyethylene
SBR reactor	1 Acrylic
Effluent tank	60 L polyethylene
Air pump	Flow rate 1 L/min
Air diffusers	-
Agitator	-
Automatic timer	-
Influent pump	Flow rate 0.52 L/min
Effluent pump	Flow rate 0.49 L/min

Table 3.3 A cycle during sequencing batch reactor (SBR) operation

Time (min)	process	Duration (min)	Volume (L)
0-8	Feed	8	6.7
0-420	Aeration + Mixing	420	10
420-480	Settle	60	10
450-480	Draw	30	6.7

2) Operating conditions and variable parameter for SBR

Operating conditions for SBR system are shown in Table 3.4.

Table 3.4 Operating conditions and variable parameters for SBR system

Control parameters		Variable parameter
Formaldehyde concentration	: optimum from batch test	SRT : 10, 30, 60 days
MLSS	: optimum from batch test	
pH	: optimum from batch test	
COD:N:P	: 200:5:1	
Dissolved oxygen	: > 2 mg/L	
HRT (8hrs/cycle and 3cycles/day)	: 12 hrs	
Temperature	: room temperature (~30°C)	

Formaldehyde concentration, initial MLSS concentration, and pH in the SBR bioreactor were obtained from batch test. The COD:N:P ratio of 200:5:1, HRT of 12 hrs, and DO of more than 2 mg/L were controlled during the operation. In the experiment, solid retention time (SRT), an important parameter in operation of the activated sludge process, was studied to determine its effects on the SBR performance. The SRT was varied at 10, 30, and 60 days by daily wasting specific amount of mixed liquor sludge. The wasting volume of mixed liquor was estimated by the Eq. 3.1 (Grady *et al.*, 1999).

$$SRT = \frac{VX}{F_w X_w} \tag{3.1}$$

where V is the effective volume of SBR (L), X is the mixed liquor suspended solid in the reactor (mg/L), F_w is the solid wasting rate (L/day), and X_w is the mixed liquor suspended solid in the waste stream reactor (mg/L).

While wasting the solids from the mixed liquor in the reactor, the value of X and X_w are identical. Eq. 3.1 may be simplified to Eq. 3.2.

$$SRT = \frac{V}{F_w} \tag{3.2}$$

According to Eq. 3.2, the volumes of waste stream to maintain the SRT of 10, 30, and 60 days are 1, 0.33, and 0.17 L/day, respectively. To investigate the SBR performance in removal of formaldehyde, samples were taken from different points including influent tank aeration tank, and effluent tank. All samples were filtered through 0.45−μm filter paper, prior to analyses for formaldehyde, TOC concentrations, MLSS in reactor and TSS in effluent.

3.3.4 Membrane bioreactor (MBR)

1) Experimental set-up

Submerged MBR each reactor was made of acrylic with an effective volume of 12-L (Fig 3.4). Membrane module, installed vertically in the screened column, consisted of polysulfone hollow fiber membranes with pore size of 6,000-30,000 daltons, and filtration area of 0.85 m^2. Air bubbles were continuously supplied through air diffusers. One was located underneath the membrane modules for scouring the membrane surface, while other two diffusers

were located elsewhere at the bottom of the tank for biomass growth and turbulence in the reactor. Influent was continuously fed into the reactor at a rate of 28.8 L/day, corresponding to hydraulic retention time (HRT) of 10 hours. Permeate was obtained by using a vacuum pump, which was controlled by a water level sensor to maintain a constant water level in the bioreactor.

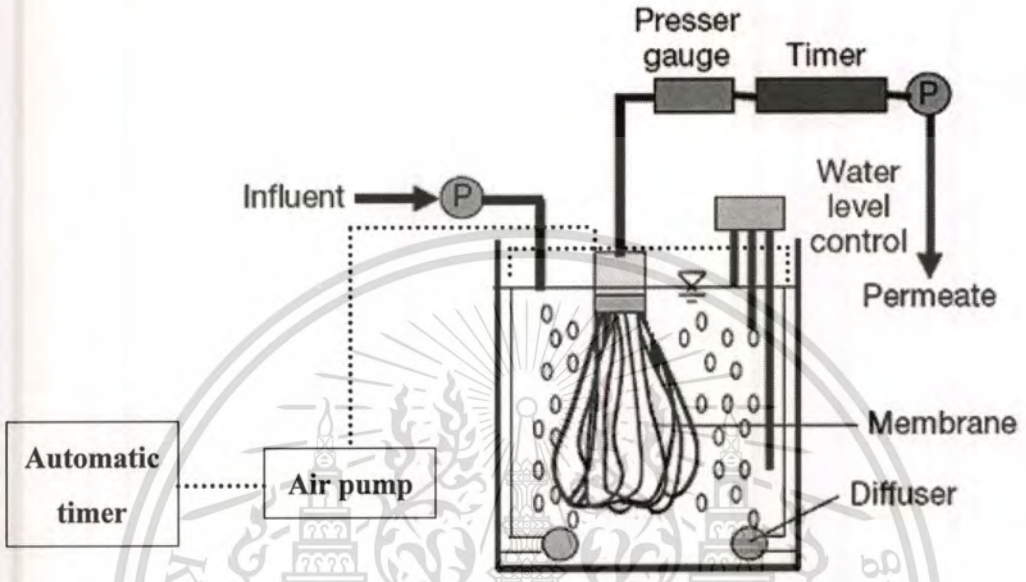


Figure 3.4 Schematic diagram of the MBR (Adapted from Lim *et al.*, 2007)

Synthetic wastewater, as described in 3.3.1, was used as feed water. Seed activated sludge, obtained from wastewater treatment plant of urea–formaldehyde resin industry, was used to inoculate the system.

2) Operating conditions and variable parameter for MBR

Operating conditions for MBR system are presented in Table 3.5.

Table 3.5 Control and variable parameters for MBR system

Control parameters		**Variable parameter**
Formaldehyde concentration	: optimum from batch test	SRT : 10, 30, 60 days
MLSS	: optimum from batch test	
pH	: optimum from batch test	
COD:N:P	: 200:5:1	
Dissolved oxygen	: > 2 mg/L	
HRT	: 10 hrs	
Temperature	: room temperature (~30°C)	

2) Operation conditions and variable parameter for MBR

The effects of SRT on the MBR performance in formaldehyde removal and membrane fouling were investigated. The SRT were varied at 10, 30, and 60 days by daily wasting mixed liquor sludge from the reactor, as mentioned in section 3.3.3. Samples were taken at different points, including permeate tank, aeration tank, and effluent tank. All samples were filtered through 0.45−μm filter paper, prior to analyses for formaldehyde and TOC concentrations.

To investigate membrane fouling, permeate flux was determined by permeate volume per membrane area per filter time. As suction pressure was not maintained constant, specific flux was reported as a function of permeate flux per suction pressure, in a unit of Liter/m^2-hr-KPa (LMH/KPa) (see example of calculation in Appendix A-2).

When membrane fouling occurred, cleaning of membrane was processed according to the following proceduces:

1) Hydrodynamic cleaning: fouled membrane was flushed with tap water for 15 minutes.

2) Caustic cleaning: the fouled membrane was placed in 0.001 M NaOH solution for 30 minutes, then rinsed with water for 15 minutes.

3) Acid cleaning: the fouled membrane was placed in 0.0001 M citric acid ($C_6H_8O_7.H_2O$) solution for 30 minutes, then rinsed with water for 15 minutes.

4) Before placing back to the MBR system, specific flux of pure water for cleaned membrane was measured.

3.3.5 Analyses and measurements

Parameters and methods of analyses that used in this study are given in Table 3.6.

Table 3.6 Measured parameters and methods

Parameters	Methods/ Equipments	References
Formaldehyde concentration	Colorimetric method, UV-Vis spectrophotometer (see example of calculation in Appendix A-3)	Nash, 1953
Mixed liquor suspended solids (MLSS)	Gravimetric method, Temperature 103-105°C	APHA, 2005
Total suspended solids (TSS)	Gravimetric method, Temperature 103-105°C	APHA, 2005
Total organic carbon (TOC)	Combustion method, Total organic carbon analyzer (TOC)	APHA, 2005
pH	pH meter	APHA, 2005
Dissolved oxygen (DO)	DO meter	APHA, 2005

CHAPTER 4

RESULTS AND DISCUSSION

This research studied factors affecting treatment of synthetic wastewater containing formaldehyde. Batch tests were conducted to study the effects of contact time, initial formaldehyde concentration, mixed liquor suspended solid (MLSS), and pH. The optimum conditions obtained from batch tests were used as operating parameters in the operation of SBR and MBR. The performances of SBR and MBR in removal of formaldehyde in synthetic wastewater were then investigated.

4.1 Batch tests

4.1.1 Effects of contact time and formaldehyde concentration on formaldehyde removal

In order to study formaldehyde biodegradation in the batch tests, the varied concentration of formaldehyde at 100, 200, 300, 400, 500, 750, and 1,000 mg/L were introduced to each bottle. MLSS concentration was approximately 500 mg/L in each bottle. The pH was controlled at 7 using phosphate buffer. Samples were taken at variaous contact times, i.e. of 6, 8, 12, 24, 36, and 48 hours.

As seen from Fig. 4.1, formaldehyde concentration in the solution reduced as contact time increased (see details in Table B-1, Appendix B). At contact time of 48 hours, formaldehyde was treated until the concentration was below 1 mg/L, which is a criterion for industrial effluent standard. It was also found that biological process with acclimated microorganism was able to treat formaldehyde at concentration of 1,000 mg/L to be lower than 1 mg/L, or more than 99.9% removal, at a contact time of 48 hours. Similar results were investigated by Glancer-Soljan *et al.*, (2001). It might be explained that the acclimated microorganisms composed of some strains of bacteria that could resist and survive in the environment contaminated with formaldehyde, for example, *Pseudomonas putida, Pseudomonas cepacia, Hansenula spp., Candida spp.* (Glancer-Soljan *et al.*, 2001). Moreover, those strains of microorganisms employed dehydrogenase enzyme, so that they could degrade formaldehyde as carbon substrate for their growth. Therefore, the concentration of formaldehyde in the solution decreased gradually.

According to the results from control experiment (as listed in Table B-1, Appendix B), it was proved that most formaldehyde was biodegraded by microorganisms in acclimated sludge.

As formaldehyde employed high solubility, high boiling point, and low Henry's law constant (as listed in Table 2.1 in Chapter 2), only few amount of formaldehyde was stripped out by the effect of aeration.

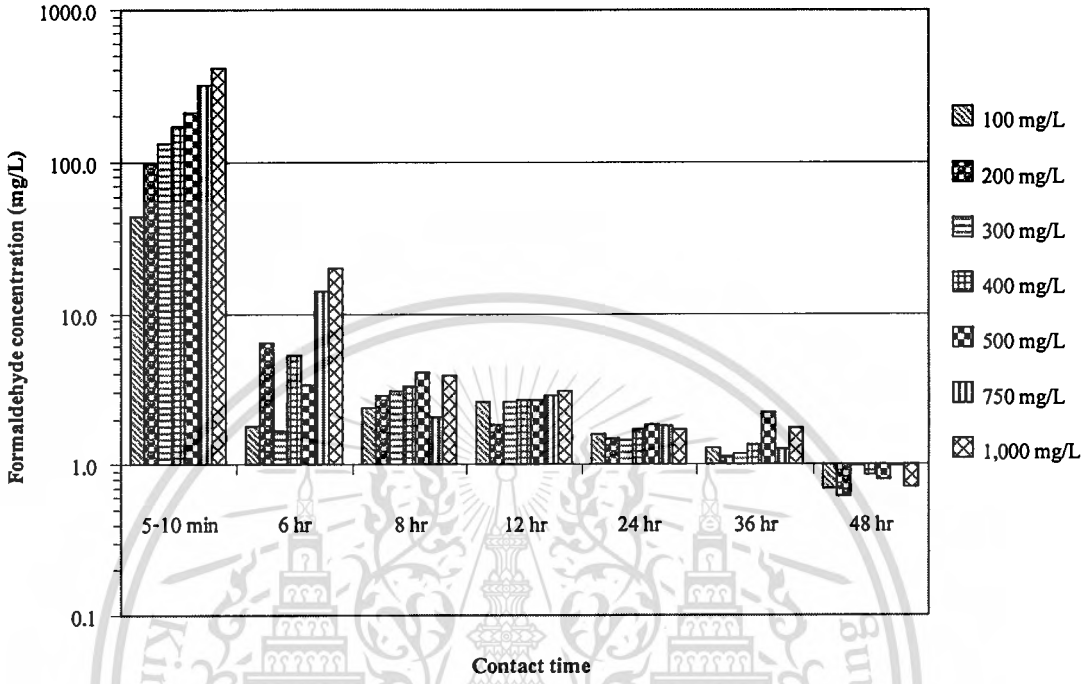


Figure 4.1 Formaldehyde concentrations remaining in effluent from batch tests at different contact times and initial formaldehyde concentrations

Although the tests showed that biological process provided high efficiency in formaldehyde removal, it was observed that with initial formaldehyde concentrations of 750 and 1,000 mg/L, the effluent appeared turbid. As seen from Fig. 4.2, TOC in the solution reduced as contact time increased. At formaldehyde concentration of 750 and 1,000 mg/L, the results of TOC analyses exhibited that organic compounds decreased slowly (see details in Table B-1 in Appendix B). It can be explained that the TOC remaining in solution originated from organic compounds of microbial cells. Some microorganisms in microbial community could not resist to high toxicity of formaldehyde so the cells became lysis and released some organic compounds into the system. The so-called soluble microbial products (SMP) caused high TOC and turbidity in the effluent (Grady *et al.*, 1999). The SMPs were utilized as substrate for other microorganisms, so that the TOC finally decreased at the contact time of 24 hours.

The results also showed that pH of the solution decreased after treatment from approximately 7 to 5 (see details in Table B-1, Appendix B). It might be explained that formic

acid was an intermediate during aerobic biodegradation of formaldehyde (Glancer-Soljan *et al.*, 2001). Buffer strength in the solution might not be strong enough to maintain the pH at 7.

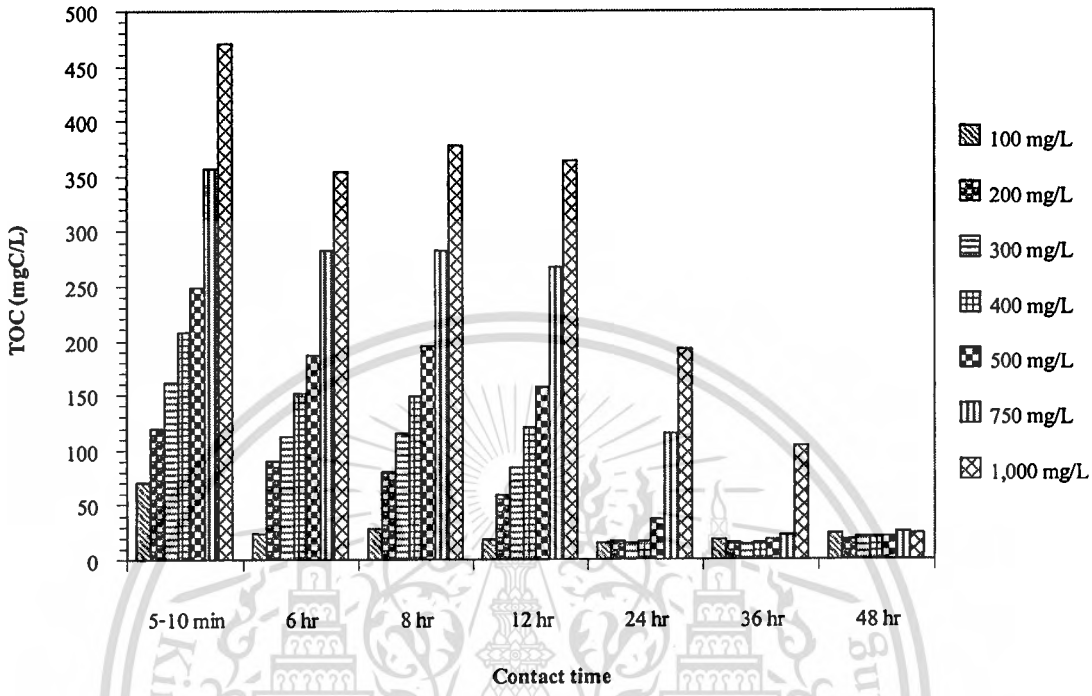


Figure 4.2 TOC concentration remaining in effluent from batch tests at different contact times and initial formaldehyde concentration

4.1.2 Effect of MLSS on formaldehyde removal

The effects of MLSS on formaldehyde removal efficiency were studied in batch tests. The MLSS were varied at 500, 1,000, 2,000, and 4,000 mg/L. Initial formaldehyde concentration was approximately 1,000 mg/L and pH of solution was maintained at 7 by using phosphate buffer. Samples were taken at contact time of 0.5, 1, 2, 4, 6, 8, and 24 hours. The results showed that MLSS concentration affected formaldehyde removal efficiency. The more MLSS content, the higher formaldehyde removal efficiency, as illustrated in Fig. 4.3 (see details in Table B-2, Appendix B). At the MLSS of 500 and 1,000 mg/L, during a period of 8 hours, formaldehyde concentration reduced gradually. Final concentration of formaldehyde in effluent was higher than 1 mg/L. As the MLSS increased, formaldehyde was removed more rapidly. With the MLSS of 2,000 and 4,000 mg/L, formaldehyde was removed from 1,000 mg/L to 1 mg/L within 2 hours and overall efficiency in formaldehyde removal was up to 99.9%.

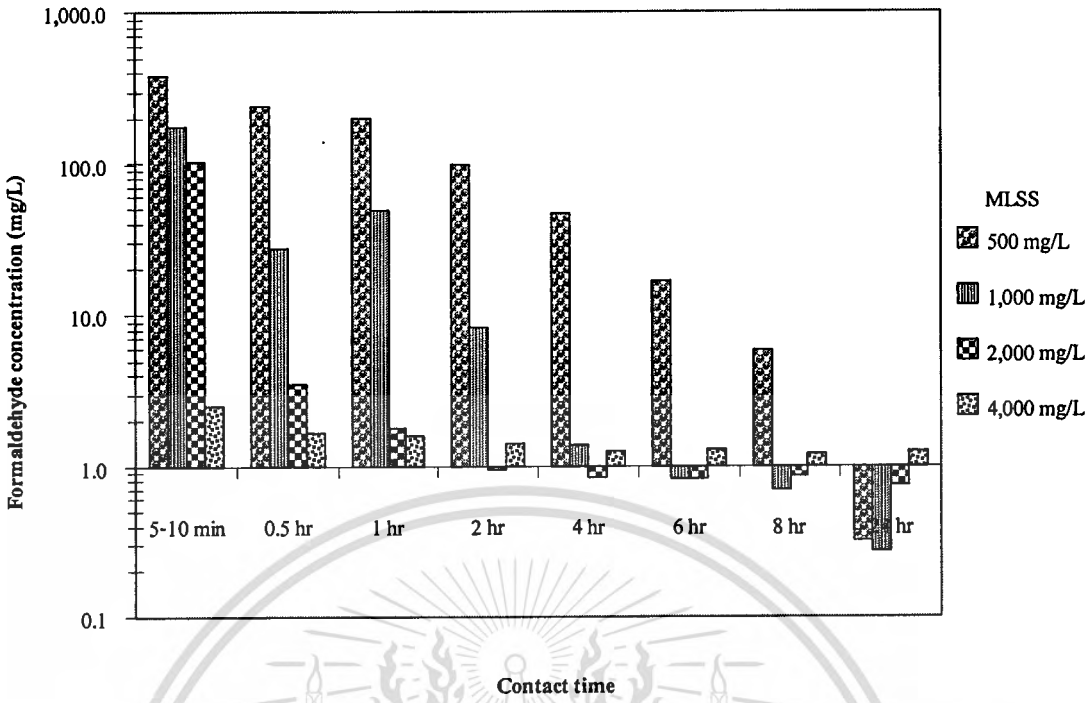


Figure 4.3 Formaldehyde concentration remaining in effluent from batch tests at different contact time and MLSS

4.1.3 Effect of pH on formaldehyde removal

The effluent of pH on formaldehyde removal was investigated in batch tests. The pH of formaldehyde containing wastewater was varied at 3, 5, 7, and 9. Initial formaldehyde concentration and MLSS were approximately 1,000 mg/L and 1,000 mg/L, respectively. As shown in Fig. 4.4, formaldehyde was removed when pH of system was 7, which was typical pH for biological processes. At this pH, formaldehyde was treated to below 1 mg/L within 8 hours of contact time. It was also found that at pH of 5, the biodegradation of formaldehyde still went on, even slower than at pH of 7. Therefore, the optimum range of pH for formaldehyde removal was in between 5 and 7. At this pH range, formaldehyde concentration was reduced from approximately 1,000 mg/L to below 1 mg/L within 24 hours (see details in Table B-3 in Appendix B).

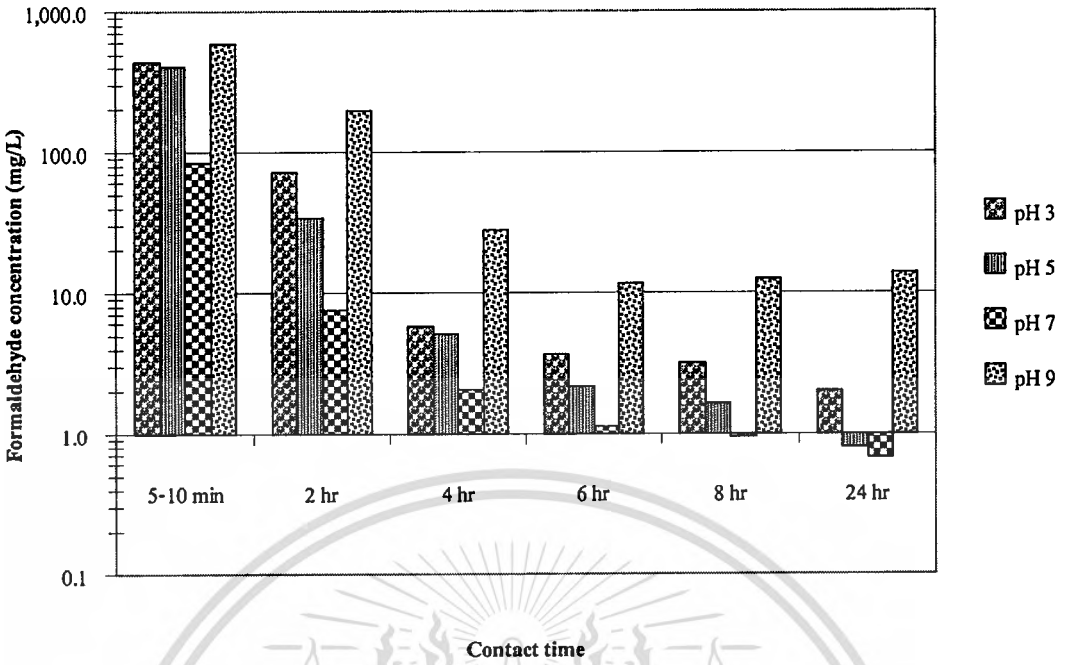


Figure 4.4 Formaldehyde concentration remaining in effluent from batch tests at different contact times and pH

4.2 Sequencing batch reactor (SBR)

4.2.1 Performance of SBR on formaldehyde removal

SBR system was operated as a cycle of 8 hrs and 3 cycles/day. Acclimated sludge was introduced into the bioreactor with an initial MLSS concentration of 1,000-2,000 mg/L. The pH was maintained at 7 with phosphate buffer. During start-up, the concentration of formaldehyde in influent was increased consistently from approximately 100 to 200, 300, 400 and 500 mg/L, then maintained at approximately 500 mg/L throughout the experiment. The results showed that the SBR exhibited an average formaldehyde removal efficiency of 99.72±0.18% for every SRT. Similar result was shown by Eiroa *et al.* (2005). Formaldehyde concentration in effluent of SBR was in a range of 0.10 and 3.01 mg/L, as shown in Fig. 4.5, with an average concentration of 1.16±0.71 mg/L (see details in Table B-4, Appendix B). The difference of formaldehyde concentrations in effluent is further explained by the effect of SRT in 4.2.2.

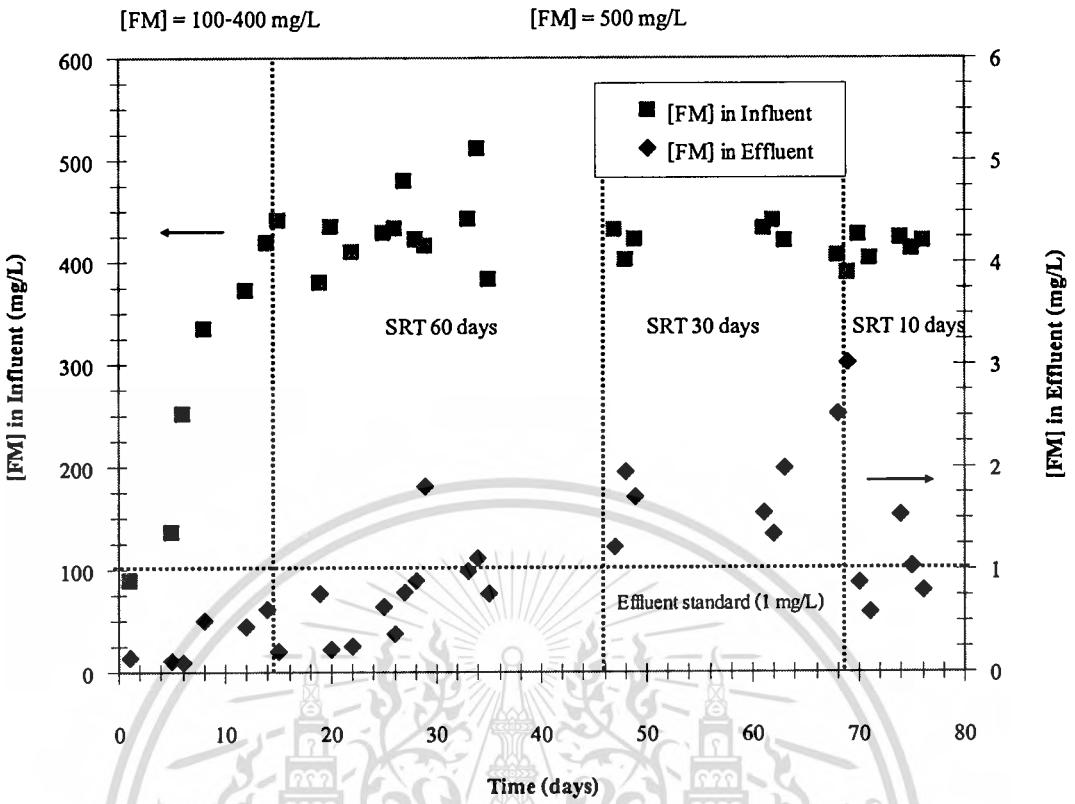


Figure 4.5 Performance of SBR on formaldehyde removal

Fig. 4.6 illustrates the TOC concentration in influent compared with the effluent TOC. The result showed that TOC of averagely 7.59 mg/L, remained in the system, even formaldehyde was removed (see details in Table B-4, Appendix B). Theoretically, 1 mg/L of formaldehyde is equivalent with 0.4 mg/L of TOC, hence, the remaining formaldehyde of 1.16 mg/L was equivalent with the TOC of 0.46 mg/L. The excess TOC might originate from microbial cells, known as soluble microbial products (SMPs) (Grady *et al.*, 1999). This finding was corresponding to the work of Shin and Kang (2003), regarding to the formation of SMPs during biological process.

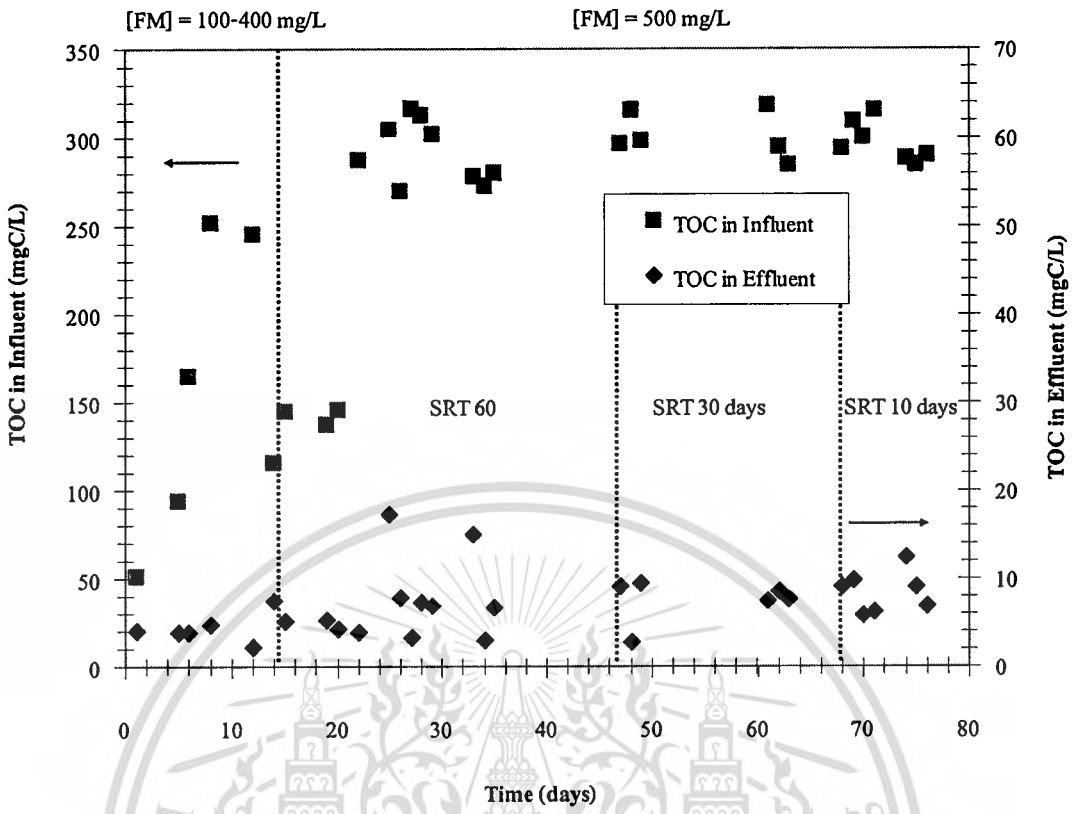


Figure 4.6 Performance of SBR on TOC removal

4.2.2 Effect of solid retention time on SBR performance

Solid retention time (SRT) is an operating parameter in biological treatment process. Figure 4.7 showed that, at the SRT of 60 days, formaldehyde was removed 99.83±0.11%, whereas at the SRT of 30 and 10 days, the SBR system exhibited percent removal of 99.62±0.08% and 99.64±0.24%, respectively. The statistical analysis by ANOVA indicated that the mean of formaldehyde removal efficiency at SRT of 60 days was significant different with those at SRT of 30 and 10 days with 95% confidence (see details in Table C-1, Appendix C). Longer SRT (60 days) provided more efficiency in formaldehyde removal than shorter SRT (30 and 10 days). This is because the amount of MLSS at longer SRT was larger than that at shorter SRT, as shown in Fig 4.8, i.e. higher amount of acclimated microorganisms.

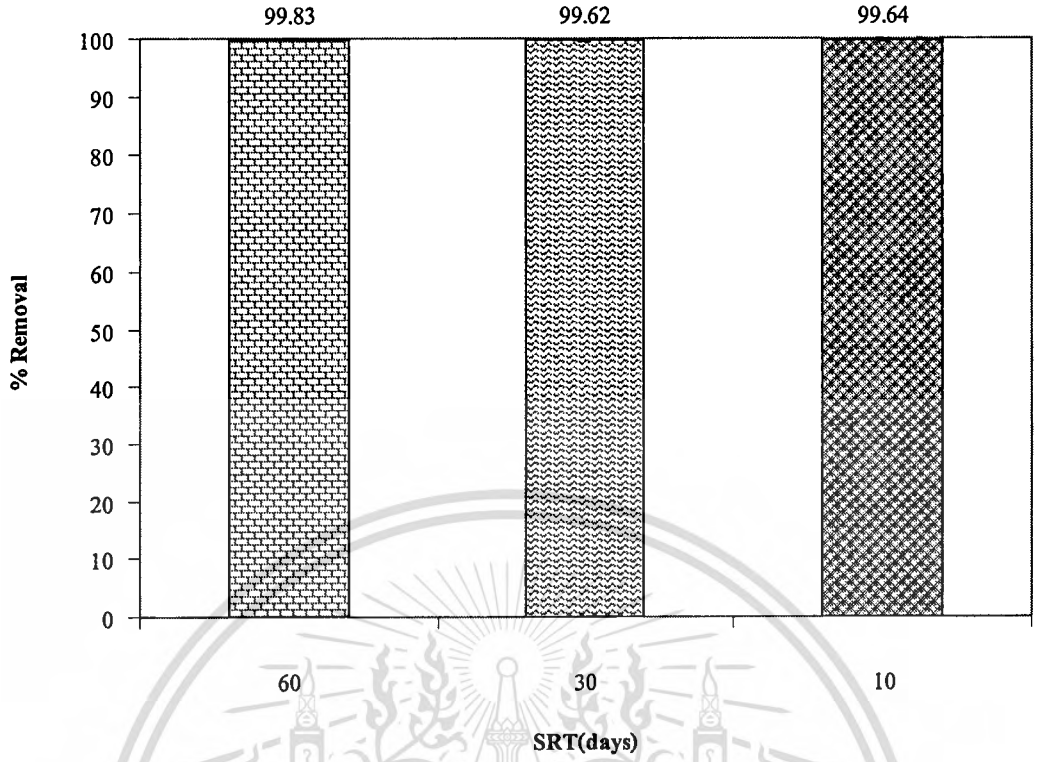


Figure 4.7 Effects of SRT on formaldehyde removal efficiency

According to Fig 4.8, it was also found that SRT affected SBR performance in terms of TSS in effluent. The result showed that an average TSS concentration in effluent at SRT of 60 days was 34 mg/L, which was below the effluent standard (50 mg/L). When decreased the SRT to 30 and 10 days, the average TSS in effluent were 90 and 127 mg/L, respectively, which exceeded the effluent standard (see details in Table B-4, Appendix B). This finding exhibited the rising trend of effluent TSS as a decrease of SRT. It can be explained that sufficient sludge aging during endogenous phase encouraged the growth of filamentous organisms that were incorporated in the floc particles. Within the floc particle, the filamentous organisms provided strength of bioflocs. While at short sludge age, acclimated bacteria dispersed in the solution due to high amount of substrate. The size of young floc particles was limited to the ability of the bacteria to stick together, causing the effluent became high level of TSS (Gerardi, 2002). Therefore, high SRT, i.e. SRT of 60 days, was an optimum condition to operate the SBR to achieve high performance in terms of formaldehyde removal efficiency and other aspects regarding to effluent quality.

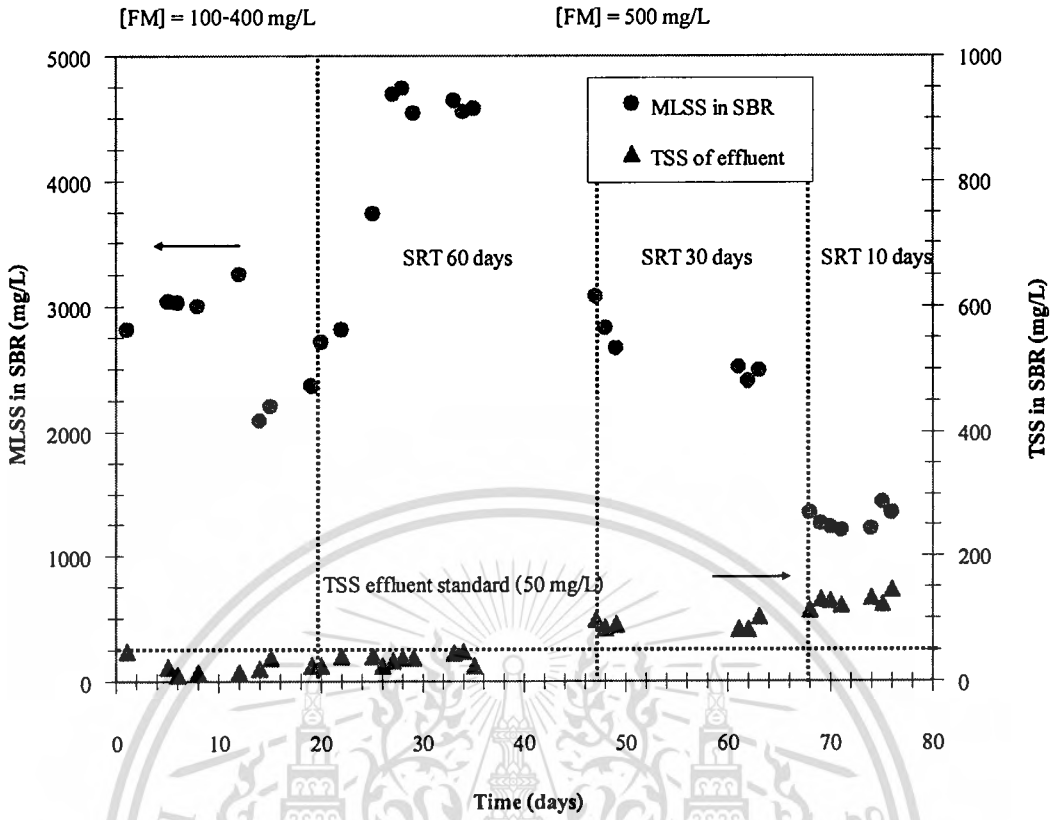


Figure 4.8 Effect of SRT on MLSS in SBR and TSS in effluent

4.3 Membrane bioreactor (MBR)

4.3.1 Performance of MBR on formaldehyde removal

The MBR was operated in semi-continuous mode with a flow rate of 28.8 L/d, and HRT of 10 hours. Acclimated sludge was introduced into the bioreactor with an initial MLSS concentration of 2,000 mg/L. When the system was started up, the concentration of formaldehyde in the influent was gradually increased from 100 to 500 mg/L, and maintained at approximately 500 mg/L. As the system had reached its steady state, MBR performance was studied. Figure 4.9 showed that the MBR reduced formaldehyde from an average concentration of 526±30 mg/L to an average concentration of 1.39±0.73 mg/L in the effluent, corresponding to removal efficiency of 99.73±0.14% (see details in Table B-6, Appendix B). This result was similar with that investigated by Eiroa *et al.* (2005). It was also found that at SRT of 60 days the MBR system provided higher efficiency than at SRT 30 and 10 days due to higher MLSS, as mentioned earlier.

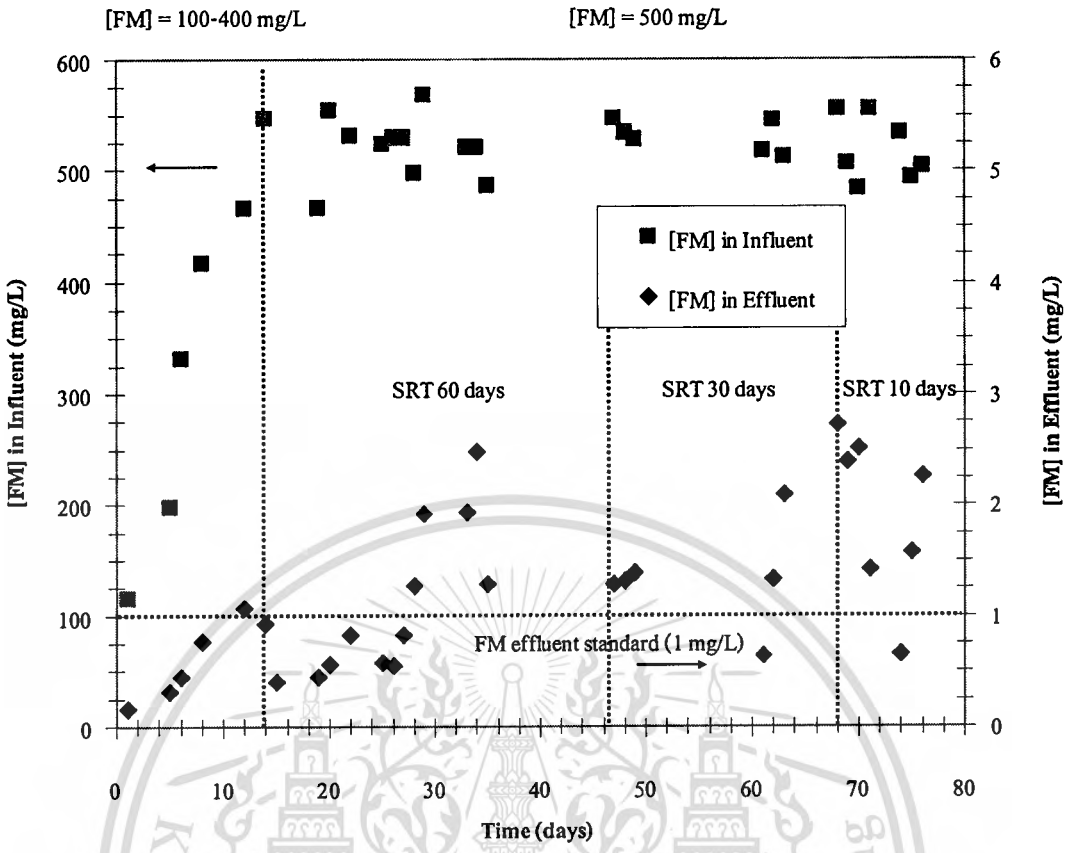


Figure 4.9 Performance of MBR on formaldehyde removal

Figure 4.10 illustrated TOC concentration which represented organic compounds in solution (see details in Table B-6, Appendix B). . As formaldehyde was used as a sole carbon source for the MBR, the TOC in the influent represented formaldehyde only. Theoretically, 1 mg/L of formaldehyde contains 0.4 mg/L of TOC, so the remaining formaldehyde of 1.39±0.73 mg/L in the MBR accounted for the TOC concentration of 0.56 mg/L. However, effluent TOC of the MBR was stable with an average concentration of 5.16±1.20 mg/L. This finding indicated that organic compounds remaining in bulk solution was not only formaldehyde but also soluble microbial product (SMPs). These compounds might be intermediates produced from substrate metabolism during biomass growth and/or organic released from biomass decay (Shin and Kang, 2003).

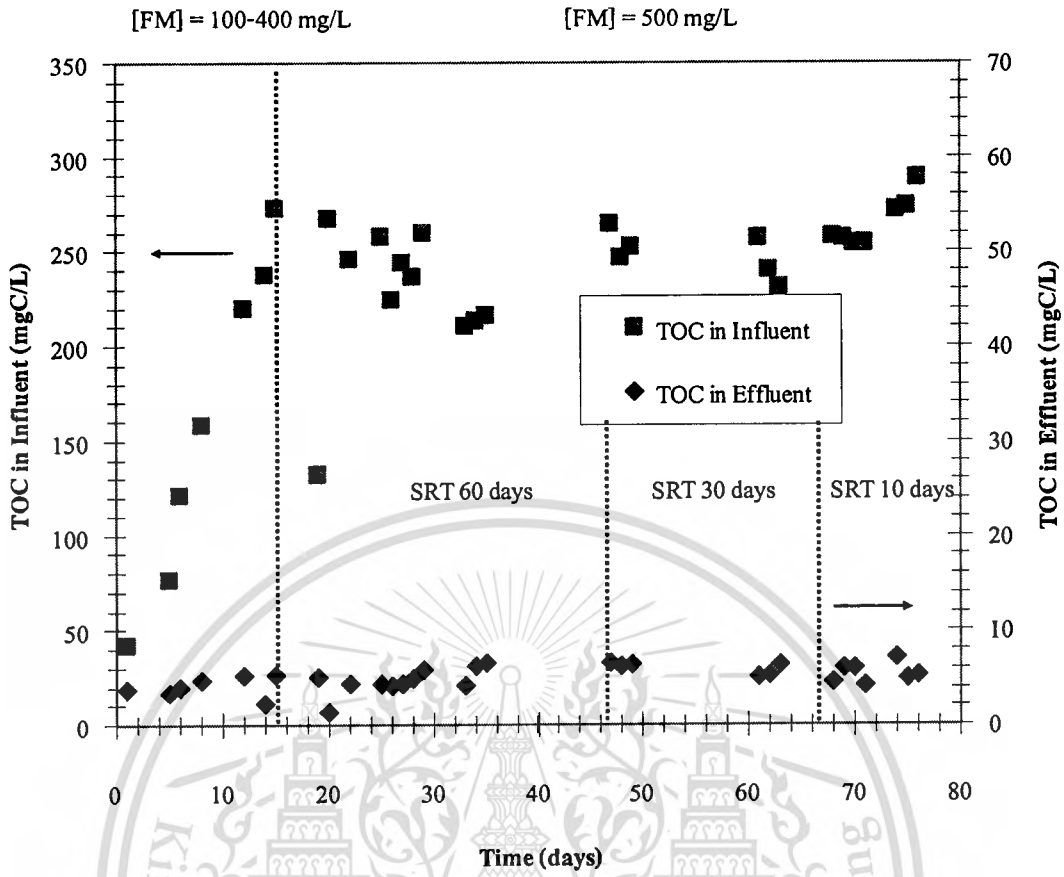


Figure 4.10 Performance of MBR on TOC removal

From the experiment, it was obvious that the MBR provided advantage in terms of solid removal. According to Fig. 4.11, The MBR exhibited good performance in TSS removal with efficiency close to 100% (see details in Table B-6, Appendix B). The effluent from MBR appeared transparent, so it might be used as raw water for reuse or recycle. Additionally, the MBR provided more stability in operation because membrane in the system could retain acclimated microorganisms.

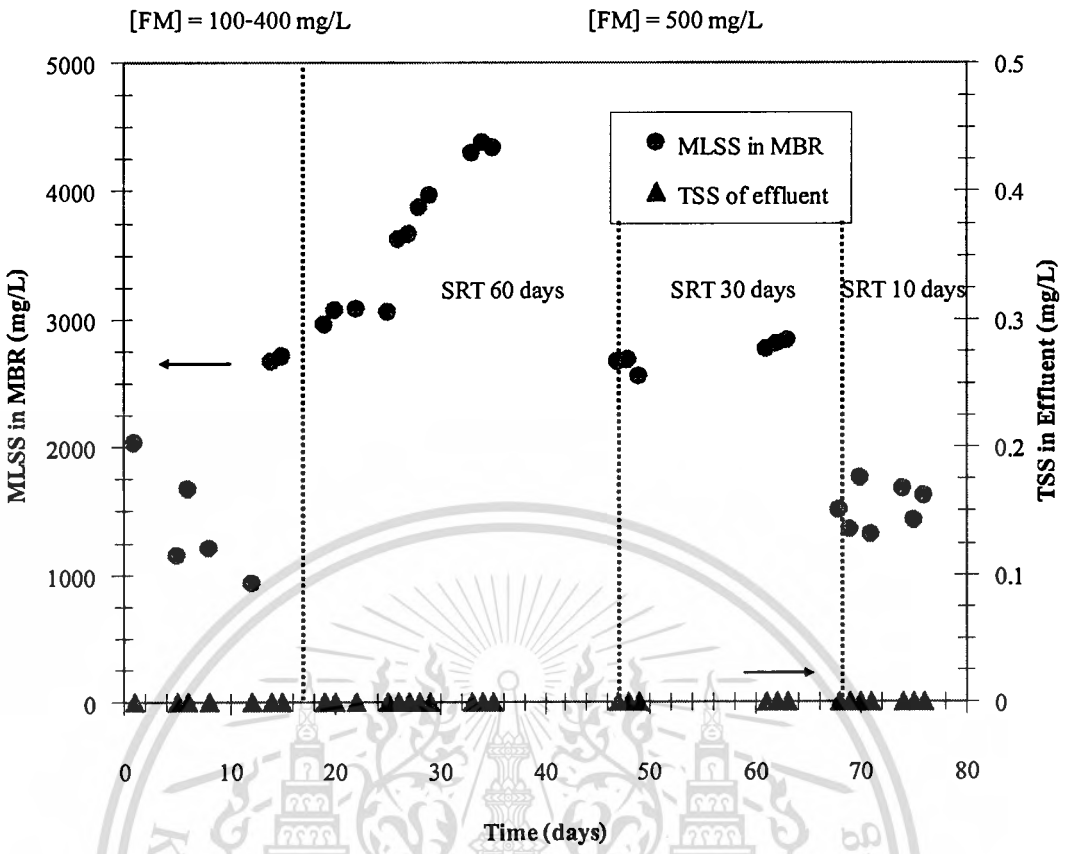


Figure 4.11 MLSS and TSS concentration in effluent of MBR system

4.3.2 Effect of solid retention time on MBR performance

The effect of SRT on formaldehyde removal by MBR was investigated. Figure 4.12 showed that formaldehyde removal efficiencies of MBR at the SRT of 60, 30, and 10 days were 99.79±0.13%, 99.75±0.09%, and 99.62±0.15%, respectively (see details in Table B-9, Appendix B). According to ANOVA result, the means of formaldehyde removal efficiency at SRT of 60 and 30 days were significant different with that at SRT of 10 days with 95% confidence (see details in Table C-1, Appendix C). The longer SRT (60 and 30 days) exhibited better performance on formaldehyde removal than the shorter SRT.

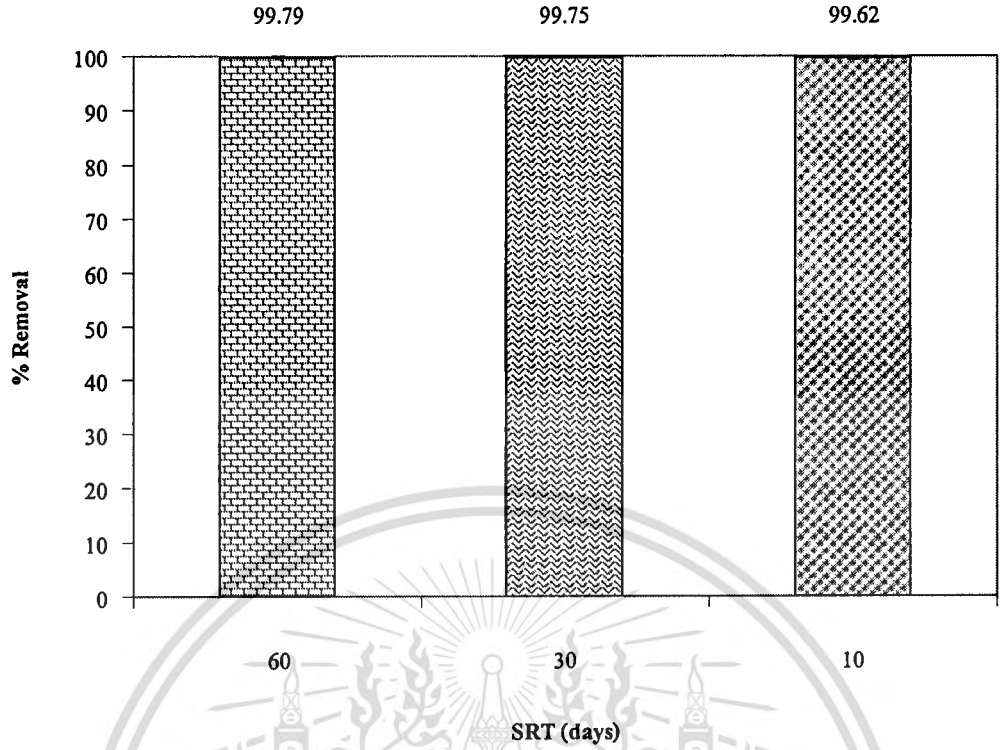


Figure 4.12 Effects of SRT on formaldehyde removal efficiency

4.3.3 Flux decline during MBR operation

Specific flux of membranes during MBR operation at varied SRTs were monitored. Figure 4.13 shows specific flux changes and cleaning cycle of the membranes. During a typical cycle (5-7 days), suspended solid accumulated on the membrane surface and the specific flux decreased (see details in Table B-8, Appendix B). The results showed similar trends of flux decline among different SRTs. This result was corresponding to those of Zoh and Stenstrom (2002) and Huang and Wu (2008). It was claimed that MLSS concentration, floc size, extracellular polymeric substances (EPS), and colloidal and soluble organic substances, were important contributors to membrane fouling. They attached to the membrane surface or clogged the membrane pores causing an increase of membrane's filtration resistance (Huang and Wu, 2008).

At the end of the cycle, increases of specific flux were the results of membrane cleaning. At SRT of 60 day, rinsing with tap water and chemical cleaning recovered 77.5% of original flux after the first cycle. Flux recovery continuously decreased to 44.6, 29.3 and 10.1%, correspondingly, for next cycles of operations (see details in Table B-8, appendix B). Flux recoveries of membranes after the cycle when operated at SRTs of 30 and 10 days were

approximately 28.9% and 30.7%, respectively. It was found that at longer sludge age (SRT of 60 days), bioflocs employed high strength due to more filamentous organisms (Zubair *et al.*, 2006). Therefore bacteria tended to stick together in biofloc and caused membrane fouling as cake formation from the bioflocs (Zoh and Stenstrom, 2002). It was removed easily by flushing with tap water, leading to high flux recovery. On the other hand, at shorter sludge age, with less MLSS, dispersed bacteria and other microorganisms might cause clogging on membrane pore as pore blockage or adsorption on membrane surface as gel formation. This phenomenon caused a difficulty in cleaning, so that, flux recovery became very low within a short period of operation.

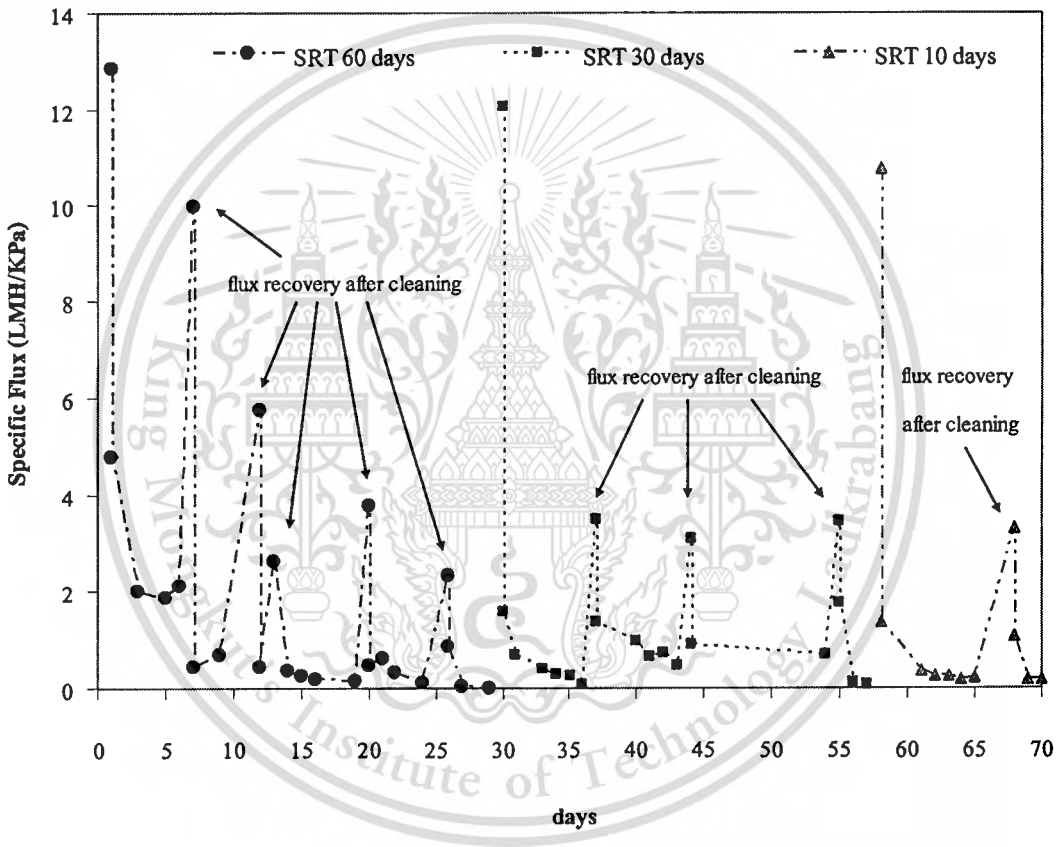


Figure 4.13 Flux decline during MBR operation at different SRTs and flux recovery after membrane cleaning

4.4 Comparison of SBR and MBR performance in formaldehyde removal

The performance of SBR and MBR was compared. Similarly, the SBR and MBR were found to be effective in formaldehyde removal, with average removal efficiency of 99.7% as shown in Fig. 4.14. The results of ANOVA indicated that there was no significant difference in the means of formaldehyde removal efficiency between SBR and MBR at every SRT with 95% confidence

(see details in Table C-3, Appendix C). Both systems functioned well in terms of formaldehyde removal. However, the MBR provided more stability in operation of the system than the SBR. The formaldehyde concentration in SBR and MBR reactors were monitored. Figure 4.15 exhibited that high formaldehyde load was fed into the SBR at the beginning of a cycle (see details in Table B-9, Appendix B). High concentration of formaldehyde might be toxic to some microorganisms in the mixed cultures. This might damage to the system in case of formaldehyde shock load. As the MBR was operated in continuous mode, formaldehyde was diluted in the reactor, leading to low formaldehyde loading. At the same time, formaldehyde was continuously degraded by microorganisms. Figure 4.16 indicated that formaldehyde concentration in MBR reactor was stable all the time of operation (see details in Table B-9, Appendix B). This finding reflected more stability in operation of the MBR system compared to the SBR.

The MBR exhibited higher benefit in terms of TSS removal, as shown in Fig. 4.17 (see details in Table B-6, Appendix B). The membrane in the MBR could retain acclimated microorganisms at all SRTs, whereas the dispersed sludge in the SBR was possibly washed out with the supernatant in case that the biofloc could not form properly. The effluent from the MBR appeared transparent without TSS, as shown in Fig. 4.18 (see details in Table B-6, Appendix B). It might be appropriate to use the effluent from the MBR as raw water for water recycle or reuse. However, limitation of the MBR was membrane fouling, leading to high energy consumption, low permeation, and cleaning cost. These aspects should be considered in selection of the system for the treatment of formaldehyde.

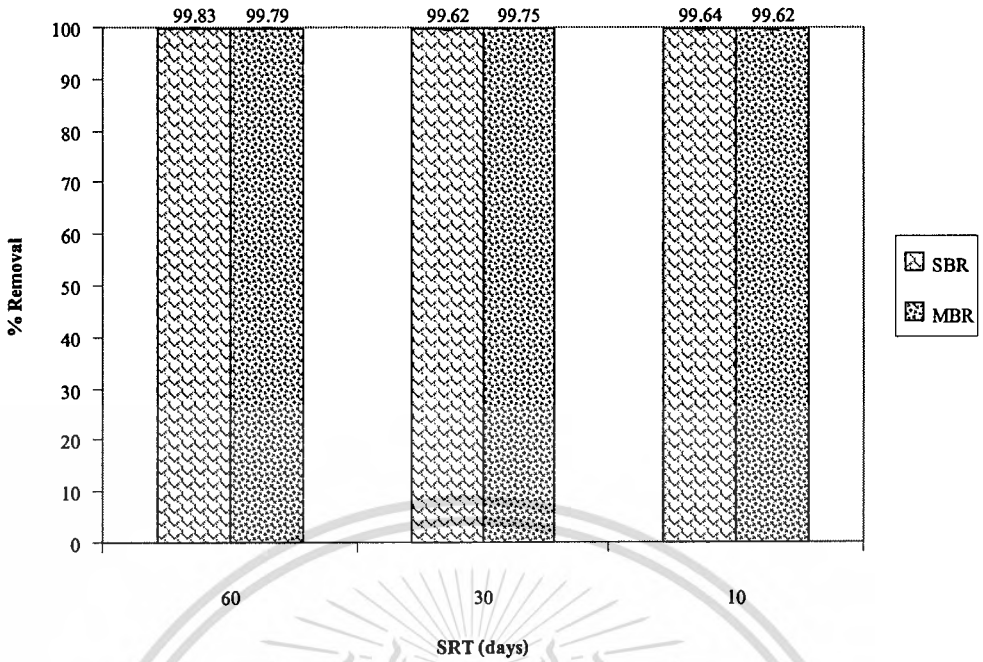


Figure 4.14 Comparison of formaldehyde removal efficiency between SBR and MBR

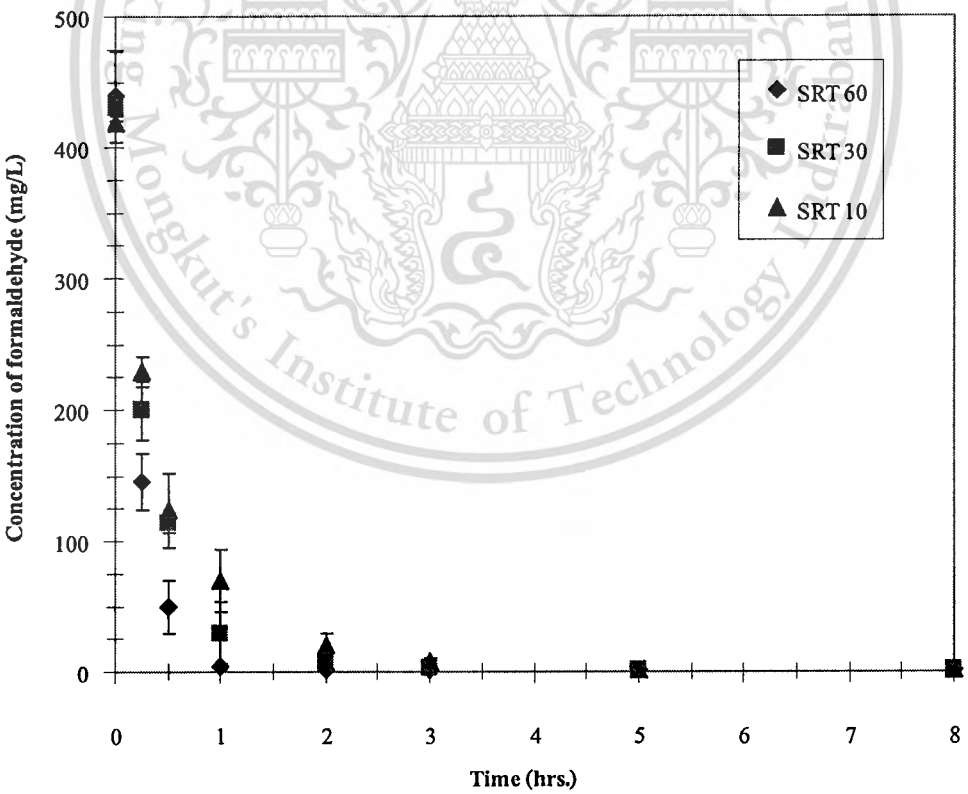


Figure 4.15 Profiles of formaldehyde concentration during an operating cycle of SBR a different SRT

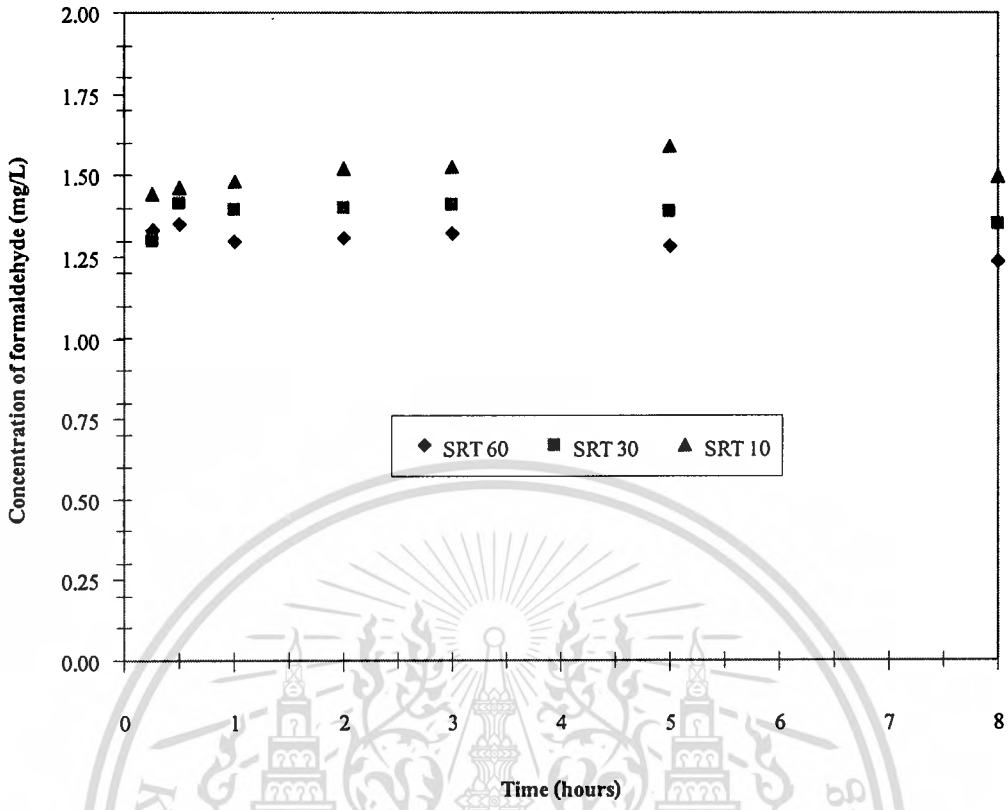


Figure 4.16 Profiles of formaldehyde concentration during an operating cycle of MBR a different SRT

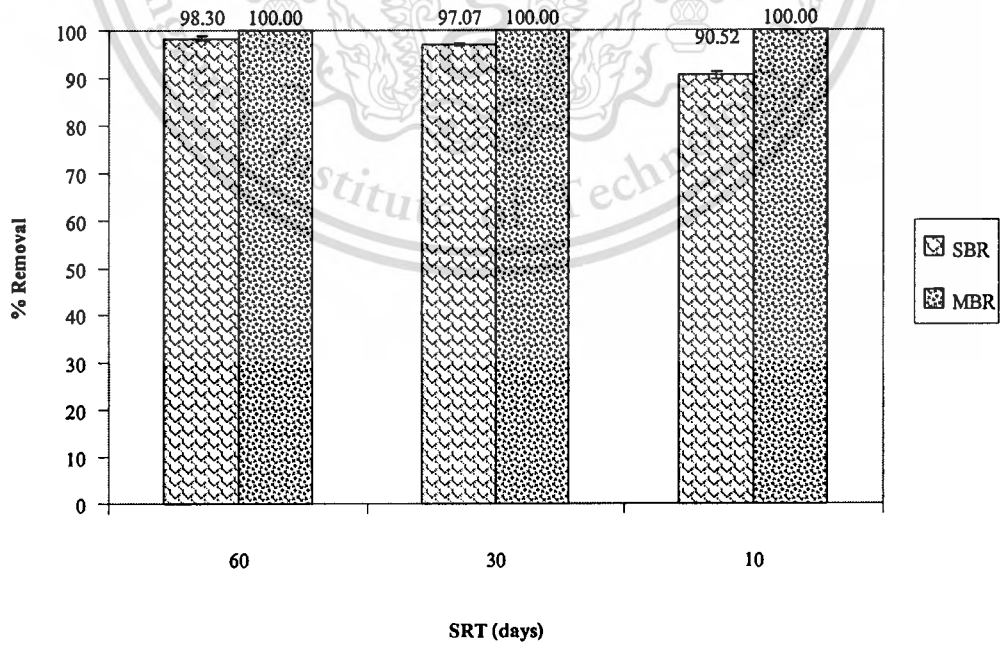


Figure 4.17 Comparison of TSS removal efficiency between SBR and MBR

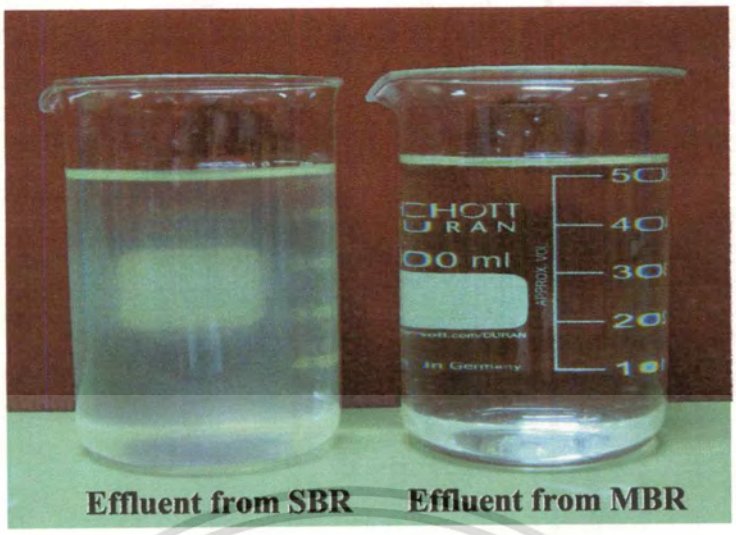


Figure 4.18 Appearance of effluents from between SBR and MBR

CHAPTER 5

CONCLUSIONS AND SUGGESTIONS

5.1 Conclusions

- Batch test

Aerobic biological process using acclimated sludge was able to treat formaldehyde. Longer contact time provided higher formaldehyde removal efficiency. The results from batch tests showed that at every initial concentration, formaldehyde was removed to below 1 mg/L at contact time of 24 and 48 hours. However, at higher formaldehyde concentration (750 and 1,000 mg/L), the effluent appeared high turbid due to high TSS in effluent. The more MLSS content, the higher formaldehyde removal efficiency. An increase of MLSS enhanced the efficiency of formaldehyde removal at shorter period of contact time. The MLSS in a range of 1,000 – 2,000 mg/L was found to remove 99.9% of formaldehyde at a contact time at least 6 hours. With the MLSS in a range of 2,000 – 4,000 mg/L, formaldehyde was removed from 1,000 mg/L to approximately 1 mg/L within 2 hours and overall efficiency in formaldehyde removal was up to 99.9%. The optimum range of pH for formaldehyde removal was in between 5 and 7. At this pH range, formaldehyde removal efficiency was 99.9% within 8 hours.

- Sequencing batch reactor (SBR)

The SBR removed formaldehyde from concentration approximately 500 mg/L to average 1.16 mg/L, corresponding to removal efficiency of 99.7%. Longer SRT (60 days) provided higher removal efficiency than shorter SRT (30 and 10 days). TOC remaining in the SBR, as known as soluble microbial products (SMP), originated during substrate metabolism and/or biomass decay. The TSS concentration in effluent at SRT of 60 days was 34 mg/L, which was lower than those at the SRT of 30 and 10 days (90 and 127 mg/L, respectively).

- Membrane bioreactor (MBR)

The MBR reduced formaldehyde from average concentration of 526 mg/L to 1.39 mg/L, corresponding to removal efficiency of 99.7%. Similar to the SBR, longer SRT provided higher removal efficiency than shorter SRT. The MBR exhibited good performance in TSS removal with efficiency close to 100%. Flux decline during MBR operation was caused by accumulation of MLSS on membrane surface. The SRT did not affect flux decline, but flux recovery after cleaning. Longer SRT (60 days) led to larger flux recovery than shorter SRTs (360 and 10 days).

- Comparison of SBR and MBR performance

The SBR and MBR were effective in formaldehyde removal with percent removal of 99.7. The MBR exhibited high benefits in terms of TSS removal and stability of the system. However, limitation of the MBR was membrane fouling, leading to high energy consumption, low permeation, and cleaning requirement. These aspects should be considered in selection of the SBR and MBR systems for formaldehyde treatment.

5.2 Suggestions

1) Higher SRT condition should be studied to treat high strength of wastewater containing formaldehyde.

2) Treatment of formaldehyde in industrial wastewater should be studied.

3) As hollow fiber membrane was potentially fouled, other types of membranes, e.g. plate and frame membrane or tubular membrane, should be investigated.

4) Intermediates of formaldehyde biodegradation, e.g. formic acid, should be analyzed to indicate the performance of the system.

5) Specific types of microorganisms in formaldehyde degradation should be cultured and identified.

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APPENDIX A

A-1 Preparation and composition of synthetic wastewater used in the experiment.

Preparation of synthetic wastewater containing formaldehyde

Synthetic wastewater were prepared by using formaldehyde as a sole carbon source. Required nutrients, e.g. nitrogen, phosphorus, and others, were added into the synthetic wastewater for microbial growth and metabolism. The examples of calculation for preparation of synthetic wastewater are described as follows:

1) Preparation of stock formaldehyde from formaldehyde solution (HCHO) 40% w/v

Required FM concentration = 10,000 mg/L

Required Volume = 1,000 mL

$$C_1V_1 = C_2V_2$$

$$400{,}000mg/L \times V_1 = 10{,}000mg/L \times 1{,}000mL$$

$$V_1 = \frac{10{,}000mg/L \times 1{,}000mL}{400{,}000mg/L}$$

$$V_1 = 25mL$$

Therefore, to prepare stock formaldehyde at a concentration of 10,000 mg/L, (40%w/v) formaldehyde of 25 mL was diluted to 1000 mL.

2) Preparation of formaldehyde solution at various concentrations from 10,000 mg/L stock formaldehyde solution.

Required FM concentration = 10,000 mg/L

Required Volume = 1,000 mL

$$C_1V_1 = C_2V_2$$

$$10{,}000mg/L \times V_1 = 1{,}000mg/L \times 1{,}000mL$$

$$V_1 = \frac{1{,}000mg/L \times 1{,}000mL}{10{,}000mg/L}$$

$$V_1 = 100mL$$

Therefore, to achieve 1,000 mg/L formaldehyde concentration, 100 mL of stock solution of 10,000 mg/L was dilution to 1,000 mg/L.

3) Ammonium sulphate $(NH_4)_2SO_4$

COD:N:P = 200:5:1 (Eiroa *et al.*, 2004)

Example: Calculation of amount of ammonium sulphate to be added in synthetic wastewater with [FM] = 1,000 mg/L.

Conversion of formaldehyde to TOC and TOC to COD

HCHO 30.03 mg has C = 12 mg

HCHO 1,000 mg has C $= \frac{12mg \times 1{,}000mg}{30.03mg} = 400$ mg

$\therefore$ TOC = 400 mg/L

$$HCHO + O_2 \longrightarrow CO_2 + H_2O$$

$$\frac{COD}{TOC} = \frac{32}{12}$$

$$\frac{COD}{TOC} = 2.67$$

$$COD = 2.67TOC$$

$$COD = 2.67 \times 400mg/L$$

$$\therefore COD = 1{,}068mg/L$$

Hence, formaldehyde concentration of 1,00 mg/L is corresponding to 1,068 mg/L of COD.

COD 200 mg/L needs N = 5 mg/L

COD 1,068 mg/L needs $N = \frac{5mg/L \times 1{,}068mg/L}{200mg/L} = 26.7$ mg/L

$(NH_4)_2SO_4$ was used as nitrogen source.

N 28 g from $(NH_4)_2SO_4$ 132 g

N 26.7 mg/L from $(NH_4)_2SO_4$ $\frac{132g \times 26.7mg/L}{28g} = 126$ mg/L

Stock $(NH_4)_2SO_4$ solution of 5,000 mg/L was used to prepare synthetic wastewater.

Required Volume 1,000 mL

$$C_1V_1 = C_2V_2$$

$$5{,}000mg/L \times V_1 = 126mg/L \times 1{,}000mL$$

$$V_1 = \frac{126mg/L \times 1{,}000mL}{5{,}000mg/L} = 25.2 \text{ mg/L}$$

Hence, to prepare N of 126 mg/L for 1L, the 5,000 mg/L $(NH_4)_2SO_4$ solution with volume of 25.2 mL was required.

4) Phosphate buffer

The pH was controlled at 7 using phosphate buffer by potassium dihydrogen phosphate (KH_2PO_4) and disodium hydrogen phosphate (Na_2HPO_4).

$$H_3PO_4 \leftrightarrow H^+ + H_2PO_4^- \qquad pKa_1 = 2.15$$

$$H_2PO_4^- \leftrightarrow H^+ + HPO_4^{2-} \qquad pKa_2 = 7.20$$

$$H_2PO_4^{2-} \leftrightarrow H^+ + PO_4^{3-} \qquad pKa_3 = 12.40$$

$$pH = pKa + \log\frac{[salt]}{[acid]}$$

$$pH = pKa + \log\frac{[HPO_4^{2-}]}{[H_2PO_4^-]} \qquad (1)$$

Required pH = 7

pKa_2 = 7.2

Total concentration = 0.01 M

$$[H_2PO_4^-] + [HPO_4^{2-}] = 0.01$$

$$[H_2PO_4^-] = 0.01 - [HPO_4^{2-}] \qquad (2)$$

Substitute (2) in (1)

$$7 = 7.2 + \log\frac{[HPO_4^{2-}]}{0.01 - [HPO_4^{2-}]}$$

$$-0.2 = \log\frac{[HPO_4^{2-}]}{0.01 - [HPO_4^{2-}]}$$

$$10^{-0.2} = \log\frac{[HPO_4^{2-}]}{0.01 - [HPO_4^{2-}]}$$

$$[HPO_4^{2-}] = 0.004 \text{ M}$$

$[HPO_4^{2-}]$ was prepared by using Na_2HPO_4 with molecular weight of 142 g/mol.

$\therefore$ $[HPO_4^{2-}]$ 0.004M was prepared by using Na_2HPO_4 (142 g/mol x 0.004 mol/L) = 568 mg/L

$$[H_2PO_4^-] = 0.006 \text{ M}$$

$[H_2PO_4^-]$ was prepared by using $[KH_2PO_4]$ with molecular weight of 136 g/mol

$[H_2PO_4^-]$ 0.006 M was prepared by using $[KH_2PO_4]$ = (136 g/mol x 0.006 mol/L) = 816 mg/L

Hence, in preparation of 0.01 M phosphate buffer to maintain pH of 7, Na_2HPO_4 568 mg/L and KH_2PO_4 816 mg/L were used.

Overall composition of synthetic wastewater used in batch tests, SBR, and MBR are listed in Table A-1, A-2, and A-3, respectively.

Table A-1 Composition of synthetic wastewater used in batch tests

Chemicals	Molecular weight (g/mol)	Amount	Influent concentration (mg/L)
Formaldehyde (HCHO)	30.03	100.00 ml/L[a]	1,000.00
Ammonium sulphate ($(NH_4)_2SO_4$)	53.50	0.13 g/L[b]	26.70 as N
Calcium chloride anhydrous ($CaCl_2.2H_2O$)	147.00	0.01 g/L	7.55 as Ca
Magnesium sulphate ($MgSO_4.7H_2O$)	246.50	0.02 g/L	24.00 as Mg
Potassium dihydrogen phosphate (KH_2PO_4)	142.00	0.84 g/L	35.82 as P
Disodium hydrogen phosphate (Na_2HPO_4)	136.00	0.54 g/L	34.70 as P

[a] Prepared from formaldehyde stock solution at a concentration of 10,000 mg/L.

[b] In case of different formaldehyde concentration, amount of ammonium sulphate should be adjusted to maintain C:N ratio of 200:1.

Table A-2 Composition of synthetic wastewater used in SBR experiment

Chemicals	Molecular weight (g/mol)	Amount	Influent concentration (mg/L)
Formaldehyde solution (HCHO) 40% w/v	30.03	1.86 ml/L	746.29
Ammonium sulphate ($(NH_4)_2SO_4$)	53.50	0.44 g/L	94.03 as N
Calcium chloride anhydrous ($CaCl_2.2H_2O$)	147.00	0.01 g/L	7.55 as Ca
Magnesium sulphate ($MgSO_4.7H_2O$)	246.50	0.02 g/L	24.00 as Mg
Potassium dihydrogen phosphate (KH_2PO_4)	142.00	0.84 g/L	35.82 as P
Disodium hydrogen phosphate (Na_2HPO_4)	136.00	0.54 g/L	34.70 as P

Table A-3 Composition of synthetic wastewater used in MBR experiment

Chemicals	Molecular weight (g/mol)	Amount	Influent concentration (mg/L)
Formaldehyde solution (HCHO) 40% w/v	30.03	1.25 ml/L	500.00
Ammonium sulphate ($(NH_4)_2SO_4$)	53.50	0.44 g/L	94.03 as N
Calcium chloride anhydrous ($CaCl_2.2H_2O$)	147.00	0.01 g/L	7.55 as Ca
Magnesium sulphate ($MgSO_4.7H_2O$)	246.50	0.02 g/L	24.00 as Mg
Potassium dihydrogen phosphate (KH_2PO_4)	142.00	0.84 g/L	35.82 as P
Disodium hydrogen phosphate (Na_2HPO_4)	136.00	0.54 g/L	34.70 as P

A-2 Example of calculation for specific flux

Flow rate (permeate volume per time) = 66.67 L/hr

Effective area of hollow fiber membrane = 0.85 m^2

Vacuum pressure = 1.80 inHg

= 1.80 inHg x 3.386388 kPa/inHg

Therefore Vacuum pressure = 6.10 kPa

$$\text{Specific Flux} = \frac{\text{Flow rate}}{\text{Effective area of membrane x vacuum pressure}}$$

$$= \frac{66.67 L/hr}{0.85 m^2 \times 6.10 kPa}$$

Therefore Specific Flux = 12.87 LMH/kPa

A-3 Determination of formaldehyde

Formaldehyde reacts with acetylacetone in a condition of excess ammonia to form yellow compound of diacetyldihydro-lutidine. Formaldehyde concentration is determined by colorimetric method.

Chemicals

1. Acetylacetone

   Dissolve 75g of ammonium acetate in distilled water, add 1.5 mL of glacial acetic acid and 1 mL of acetyl acetone, then make up with distilled water to 500 mL.

2. Sodium sulfite solution

   Dissolve 125 g of anhydrous sodium sulfite in 1 L of distilled water.

3. 5% Sulfuric acid

   Add 5 mL of 98% (w/w) sulfuric acid or 36N sulfuric acid in 95 mL distilled water.

4. 1 N Sulfuric acid
5. 1N Sodium hydroxide
6. Thymolphthalein indicator solution

   Dissolve 1 g of thymolphthalein in 1L of 98% ethanol (v/v)

Procedure

1. **Standard curve**

   1.1 Prepare 10,000 mg/L of stock formaldehyde solution by 25 mL of 38-40% w/v formaldehyde diluted to 1 L by using distilled.

   1.2 Pipet 25 mL of sodium sulfite solution into a flask, add a few drops of thymolphthalein indicator (the solution turns blue), then add 1-2 drops of 1N sulfuric acid until the blue solution becomes transparent.

   1.3 Add 25 mL of formaldehyde solution into the flask (the solution turns blue), then titrate with 1N sulfuric acid. At the end point, the solution becomes transparent.

   1 mL of 1 N sulfuric acid equivalent with 30.03 mL of formaldehyde. The exact formaldehyde concentration is determined by the following equation.

$$\text{Formaldehyde concentration (mg/L)} = \frac{A \times 30.03 \times 1000}{25}$$

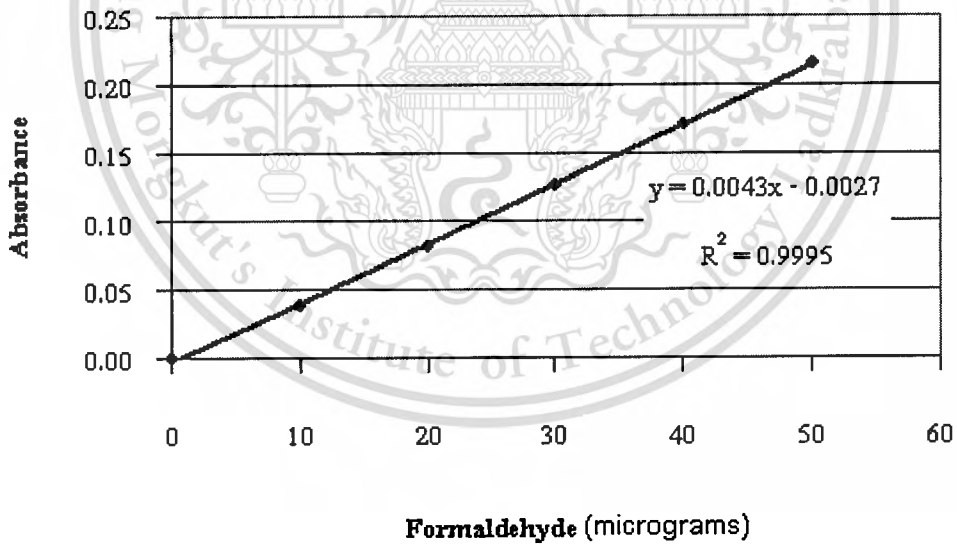
where A = volume of 1 N sulfuric acid used for titration

1.4 Prepare 10 mg/L formaldehyde solution using the solution from 1.3 (known concentration). Dilute 10 mg/L of formaldehyde to obtain a set of standard solution containing contents of formaldehyde of 10, 20, 30, 40 and 50 micrograms by pipetting 10 mg/L formaldehyde at volume of 1, 2, 3, 4 and 5 mL, respectively. Distilled water is used as a blank.

1.5 Add standard solutions and blank in 25-mL volumetric flasks, then add 5mL of 1N sulfuric acid in each flask and mix thoroughly.

1.6 Add 5 mL of 1N sodium hydroxide and adjust the volume to 25 mL with distilled water in every flask.

1.7 Pour the content from each flask into 50-mL volumetric flask, rinse with acetylacetone, then adjust the volume to 50 mL with acetylacetone. Heat up each flask at 60°C for 10 minutes. Let the flask cool down before measuring absorbace at a wavelength of 425 nm. Standard curve is plotted between formaldehyde content (in microgram) and absorbance.



2. Sample analysis

1. Pipet 15 mL of sample into a 50-mL volumetric flask.
2. Add 5mL of 1N sulfuric acid in the flask and mix thoroughly.
3. Add 5 mL of 1N sodium hydroxide

4. Adjust the volume to 50 mL with acetylacetone. Heat up the flask at 60°C for 10 minutes. Let the flask cool down before measuring absorbance at a wavelength of 425 nm.
5. Content of formaldehyde in sample is achieved when compared the absorbance with standard curve. Formaldehyde concentration in sample is determined by the following equation:

$$\text{Formaldehyde (mg/L)} = \frac{\text{Formaldehyde content (micrograms)}}{\text{Sample volume (mL)}^{*}}$$

*Sample volume equals to 15 mL.

APPENDIX B

Experimental Data

Table B-1 Effects of formaldehyde concentration on formaldehyde removal efficiency at different at contact time

Sample	5-10 minutes			6 hours			8 hours		
	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH
control-100 mg/L	75.56	33.89	7.0	105.27	47.54	7.0	97.05	44.94	7.2
control-200 mg/L	218.29	96.26	7.0	217.26	91.84	7.1	194.09	85.38	7.0
control-300 mg/L	303.26	138.50	7.0	310.49	134.60	7.2	283.13	129.50	7.1
control-400 mg/L	403.51	181.30	7.0	426.25	178.70	7.0	371.01	175.80	7.1
control-500 mg/L	535.81	241.20	7.0	527.55	219.00	7.0	480.10	215.50	7.2
control-750 mg/L	778.71	350.00	7.0	788.01	331.70	7.0	726.57	325.40	7.0
control-1000 mg/L	1038.14	458.80	7.0	1040.21	443.40	7.1	933.64	433.30	7.1
FM-100 mg/L	43.69	70.35	6.1	1.80	24.05	5.5	2.40	27.45	5.5
FM-200 mg/L	97.26	119.00	6.5	6.34	90.33	5.4	2.84	79.53	5.2
FM-300 mg/L	130.85	162.10	5.5	1.67	111.70	5.2	3.09	114.50	5.1
FM-400 mg/L	171.78	208.40	5.5	5.22	151.30	5.0	3.33	148.90	5.0
FM-500 mg/L	212.09	249.20	5.5	3.36	187.10	5.0	4.09	194.90	5.0
FM-750 mg/L	315.66	356.10	5.2	13.92	281.80	5.4	2.06	282.80	5.1
FM-1000 mg/L	414.88	470.70	5.9	19.92	353.80	5.2	3.88	378.00	5.2

Table B-1 (Cont.) Effects of formaldehyde concentration on formaldehyde removal efficiency at different at contact time

Sample	12 hours			24 hours			36 hours			48 hours		
	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH
control-100 mg/L	101.59	45.73	7.2	104.37	44.52	7.0	74.29	41.85	7.2	13.57	20.40	7.2
control-200 mg/L	201.67	91.83	7.0	217.83	88.74	7.1	196.64	86.96	7.0	133.20	84.99	7.0
control-300 mg/L	291.21	134.10	7.0	321.52	131.10	7.2	274.42	130.60	7.1	200.26	127.20	7.0
control-400 mg/L	379.09	179.60	7.0	425.56	177.60	7.0	391.21	174.70	7.1	420.16	171.50	7.0
control-500 mg/L	471.01	221.80	7.1	534.65	217.30	7.0	527.65	214.10	7.2	537.98	205.20	7.1
control-750 mg/L	726.57	341.70	7.0	809.39	326.40	7.0	777.78	323.10	7.0	807.75	311.60	7.0
control-1000 mg/L	939.70	443.80	7.1	1055.86	430.00	7.1	993.80	425.90	7.1	1021.71	410.00	7.1
FM-100 mg/L	2.64	18.92	5.5	1.62	16.09	5.0	1.30	18.79	5.5	0.69	24.19	5.3
FM-200 mg/L	1.86	58.32	5.3	1.49	16.16	5.1	1.12	14.93	5.2	0.61	17.69	5.5
FM-300 mg/L	2.63	83.70	5.2	1.46	15.26	5.2	1.17	14.73	5.1	0.98	21.32	5.1
FM-400 mg/L	2.66	120.40	5.2	1.73	17.43	5.4	1.35	15.68	5.3	0.84	20.69	5.3
FM-500 mg/L	2.67	157.30	5.0	1.85	36.80	5.0	2.22	18.60	5.2	0.79	20.99	5.1
FM-750 mg/L	2.84	266.90	5.1	1.82	115.10	5.3	1.27	21.93	5.1	0.97	25.32	5.4
FM-1000 mg/L	3.09	364.30	5.2	1.71	192.00	5.2	1.76	104.60	5.2	0.71	23.34	5.2

Table B-2 Effects MLSS on formaldehyde removal efficiency at different at contact time

Sample	5-10 minutes			0.5 hours			1 hours			2 hours		
	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH
control	1117.45	404.10	7.1	1018.16	396.60	7.3	1018.16	396.60	7.3	981.28	395.40	7.2
MLSS-500 mg/L	382.77	392.40	4.7	198.64	347.50	4.0	198.64	347.50	4.0	97.94	324.40	3.8
MLSS-1000 mg/L	175.51	393.80	5.9	48.87	315.00	5.0	48.87	315.00	5.0	8.35	282.40	4.5
MLSS-2000 mg/L	103.23	403.40	5.4	1.79	287.80	5.3	1.79	287.80	5.3	0.95	246.20	5.5
MLSS-4000 mg/L	2.49	370.50	5.5	1.59	298.70	5.5	1.59	298.70	5.5	1.40	248.40	4.7

Table B-2 (cont.) Effects MLSS on formaldehyde removal efficiency at different at contact time

Sample	4 hours			6 hours			8 hours			24 hours		
	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH
control	984.11	397.50	7.1	1052.20	383.20	7.3	1044.63	392.60	7.2	992.62	399.90	7.0
MLSS-500 mg/L	46.51	322.10	4.7	16.63	324.00	4.7	5.80	304.50	5.4	0.32	316.60	4.7
MLSS-1000 mg/L	1.37	272.50	5.5	0.82	298.40	5.3	0.70	292.70	5.2	0.27	195.50	5.4
MLSS-2000 mg/L	0.84	249.90	5.1	0.82	288.60	5.4	0.86	258.30	5.4	0.75	15.70	4.7
MLSS-4000 mg/L	1.26	250.90	4.9	1.28	198.20	5.6	1.18	74.71	5.5	1.25	14.69	5.0

Table B-3 Effects pH on formaldehyde removal efficiency at different at contact time

Sample	5-10 minutes			2 hours			4 hours		
	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH
control pH 3	996.37	467.00	3.0	991.84	459.10	3.3	998.49	420.20	3.3
control pH 5	1021.84	399.90	5.1	1012.39	395.60	5.2	960.38	384.50	4.6
control pH 7	969.83	393.60	7.0	999.15	390.20	6.4	958.49	312.90	6.4
control pH 9	829.48	449.30	9.0	834.01	449.80	7.4	894.47	421.30	7.5
pH 3	442.86	436.20	3.0	73.03	433.80	3.2	5.76	368.40	3.2
pH 5	404.54	349.60	5.0	34.19	329.30	4.3	5.09	314.00	4.5
pH 7	84.54	333.50	7.0	7.63	324.30	5.8	2.06	298.50	6.2
pH 9	597.96	428.50	9.0	194.33	430.10	7.3	27.80	330.60	7.5

Table B-3 (cont.) Effects pH on formaldehyde removal efficiency at different at contact time

Sample	6 hours			8 hours			24 hours		
	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	FM (mg/L)	TOC (mgC/L)	pH
control pH 3	1023.07	418.00	3.4	992.31	417.70	3.3	995.09	403.20	3.3
control pH 5	979.29	359.10	4.3	1011.44	363.90	4.7	983.07	352.00	5.3
control pH 7	960.38	361.00	6.4	1003.88	360.00	6.4	977.40	344.00	6.2
control pH 9	850.02	422.50	7.5	794.17	416.80	7.5	816.39	404.00	7.5
pH 3	3.68	379.80	3.3	3.16	403.00	3.3	2.01	355.30	3.2
pH 5	2.17	317.40	3.2	1.63	231.70	4.5	0.79	266.50	3.2
pH 7	1.14	220.40	5.7	0.95	144.60	5.9	0.67	75.54	6.5
pH 9	11.72	368.70	7.4	12.56	317.40	7.2	13.74	262.80	7.1

Table B-4 Performance of SBR on formaldehyde removal

Date	Day	SRT (days)	Influent			SBR Reactor		Effluent				% FM Removal
			FM (mg/L)	TOC (mgC/L)	pH	MLSS (mg/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	TSS (mg/L)	
20/2/2009	1	-	133.36	51.06	7.4	2816	7.2	0.14	4.07	5.6	48	99.84
24/2/2009	5	-	203.07	93.63	7.1	3040	6.8	0.12	3.76	4.7	22	99.92
25/2/2009	6	-	376.30	164.30	7.1	3032	6.8	0.10	3.90	4.7	10	99.96
27/2/2009	8	-	498.44	252.20	7.1	3006	6.7	0.49	4.66	5.4	16	99.85
3/3/2009	12	-	554.95	245.60	7.0	3250	6.9	0.44	2.19	5.6	14	99.88
5/3/2009	14	-	625.67	115.50	7.2	2080	7.0	0.61	7.43	5.5	20	99.85
6/3/2009	15	60	658.00	144.30	7.0	2194	7.1	0.21	5.11	5.7	38	99.95
10/3/2009	19	60	565.73	137.20	6.9	2368	6.7	0.76	5.27	5.5	24	99.80
11/3/2009	20	60	648.30	145.90	6.8	2708	6.8	0.22	4.10	4.9	24	99.95
13/3/2009	22	60	610.96	287.20	7.0	2810	7.1	0.25	3.77	5.7	40	99.94
16/3/2009	25	60	638.20	305.00	7.1	3730	7.0	0.63	17.35	4.7	40	99.85
17/3/2009	26	60	645.77	269.60	7.1	5260	6.7	0.38	7.88	4.8	26	99.91
18/3/2009	27	60	715.19	316.70	7.0	4682	7.1	0.79	3.25	5.2	32	99.84
19/3/2009	28	60	630.39	312.50	7.2	4742	7.0	0.88	7.24	4.9	38	99.79

Date	Day	SRT (days)	Influent			SBR Reactor		Effluent				% FM Removal
			FM (mg/L)	TOC (mgC/L)	pH	MLSS (mg/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	TSS (mg/L)	
20/3/2009	29	60	619.25	301.60	7.0	4536	7.2	1.82	6.87	4.8	38	99.56
24/3/2009	33	60	659.56	278.60	7.1	4630	7.1	0.99	14.92	5.0	44	99.78
25/3/2009	34	60	762.83	273.00	7.1	4548	6.9	1.11	2.97	4.8	48	99.78
26/3/2009	35	60	571.27	280.20	7.2	4570	6.8	0.76	6.77	4.9	24	99.80
7/4/2009	47	30	643.62	296.00	7.0	3082	7.0	1.21	9.16	5.2	98	99.72
8/4/2009	48	30	599.09	315.30	6.8	2824	7.1	1.96	2.71	5.4	86	99.51
9/4/2009	49	30	628.18	298.00	6.7	2668	6.8	1.70	9.42	5.6	90	99.60
21/4/2009	61	30	644.34	317.90	7.1	2510	7.0	1.55	7.47	5.5	82	99.64
22/4/2009	62	30	655.96	295.00	7.3	2402	6.8	1.34	8.49	4.5	84	99.70
23/4/2009	63	30	626.46	285.00	7.2	2484	6.9	1.98	7.68	4.7	102	99.53
28/4/2009	68	10	604.96	293.80	7.0	1344	7.2	2.52	9.09	4.9	112	99.38
29/4/2009	69	10	580.16	308.70	6.9	1256	7.0	3.01	9.90	5.0	130	99.23
30/4/2009	70	10	635.88	300.10	7.2	1226	7.1	0.88	5.73	4.6	128	99.79
1/5/2009	71	10	601.27	315.30	7.1	1212	6.8	0.58	6.17	5.1	120	99.86
4/5/2009	74	10	631.43	288.10	6.8	1214	6.8	1.54	12.39	5.2	134	99.64
5/5/2009	75	10	614.88	284.10	6.8	1426	7.0	1.03	9.18	5.6	122	99.75
6/5/2009	76	10	627.29	289.80	7.2	1344	7.3	0.80	6.88	5.8	146	99.81

Table B-5 Effects of solid retention time on SBR performance

Time (hours)	FM (mg/L)						TOC (mgC/L)					
	SRT 60 days		SRT 30 days		SRT 10 days		SRT 60 days		SRT 30 days		SRT 10 days	
	Average	SD	Average	SD	Average	SD	Average	SD	Average	SD	Average	SD
0	438.96	35.09	430.73	9.99	418.78	5.62	310.27	8	299.30	17	287.33	3
0.25	145.33	21.88	199.71	22.11	229.35	11.24	144.00	24	167.70	8	186.30	24
0.5	49.54	20.54	113.90	7.33	123.60	28.07	126.83	19	150.93	2	176.00	23
1	3.67	2.12	28.86	24.04	69.44	23.88	106.73	22	118.30	15	152.20	25
2	1.58	0.92	5.92	4.52	20.43	8.78	82.98	21	76.20	12	110.11	16
3	1.26	0.76	2.26	0.36	7.02	3.54	55.18	9	37.15	10	77.01	18
5	0.99	0.66	1.71	0.39	1.43	0.47	11.74	10	8.98	1	12.94	3
8	0.68	0.41	1.60	0.36	1.13	0.39	5.66	2	7.11	1	9.48	3

Table B-6 Performance of MBR on formaldehyde removal

Date	Day	SRT (days)	Influent			MBR Reactor		Effluent				% FM Removal
			FM (mg/L)	TOC (mgC/L)	pH	MLSS (mg/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	TSS (mg/L)	
20/2/2009	1	-	115.18	41.98	7.3	2028	6.9	0.16	3.69	5.4	0	99.86
24/2/2009	5	-	197.78	76.06	7.4	1140	6.6	0.31	3.36	4.6	0	99.84
25/2/2009	6	-	331.36	120.70	7.4	1674	6.8	0.44	3.83	4.4	0	99.87
27/2/2009	8	-	416.22	158.50	7.3	1208	6.8	0.76	4.57	5.1	0	99.82
3/3/2009	12	-	466.06	219.80	7.1	922	7.0	1.06	5.12	4.9	0	99.77
5/3/2009	14	-	546.78	237.10	7.0	2664	6.9	0.92	2.19	4.8	0	99.83
6/3/2009	15	60	605.78	272.90	7.2	2708	6.8	0.41	5.24	5.2	0	99.93
10/3/2009	19	60	465.98	132.10	7.1	2952	7.1	0.45	5.01	5.1	0	99.90
11/3/2009	20	60	554.47	267.40	7.3	3070	7.2	0.56	1.28	4.8	0	99.90
13/3/2009	22	60	530.96	245.30	6.8	3084	7.1	0.82	4.26	4.9	0	99.85
16/3/2009	25	60	522.84	257.70	6.9	3054	6.7	0.58	4.30	5.9	0	99.89
17/3/2009	26	60	529.46	223.90	7.0	3614	6.8	0.55	4.08	5.5	0	99.90
18/3/2009	27	60	529.68	243.90	7.1	3656	7.0	0.82	4.34	5.3	0	99.84
19/3/2009	28	60	498.21	236.30	6.9	3872	6.9	1.27	4.92	5.1	0	99.75
20/3/2009	29	60	567.75	259.50	6.8	3968	7.1	1.91	5.81	4.6	0	99.66

Date	Day	SRT (days)	Influent			MBR Reactor		Effluent				% FM Removal
			FM (mg/L)	TOC (mgC/L)	pH	MLSS (mg/L)	pH	FM (mg/L)	TOC (mgC/L)	pH	TSS (mg/L)	
24/3/2009	33	60	521.28	210.70	6.9	4294	7.0	1.93	4.15	4.8	0	99.63
25/3/2009	34	60	520.32	212.80	7.0	4382	6.8	2.47	6.10	4.9	0	99.52
26/3/2009	35	60	486.51	216.30	6.9	4332	7.1	1.29	6.47	4.5	0	99.74
7/4/2009	47	30	546.46	264.10	6.8	2662	7.2	1.29	6.53	4.9	0	99.76
8/4/2009	48	30	533.43	246.40	7.0	2684	7.1	1.31	6.14	5.1	0	99.75
9/4/2009	49	30	528.18	252.40	7.2	2556	6.8	1.38	6.25	4.8	0	99.74
21/4/2009	61	30	517.07	257.20	7.1	2762	6.7	0.64	4.99	4.9	0	99.88
22/4/2009	62	30	545.86	239.80	6.9	2802	7.3	1.32	5.20	5.0	0	99.76
23/4/2009	63	30	511.73	231.30	6.7	2834	6.9	2.09	6.24	5.4	0	99.59
28/4/2009	68	10	555.35	258.10	7.2	1510	7.1	2.73	4.55	5.7	0	99.51
29/4/2009	69	10	505.74	257.00	7.3	1360	7.2	2.39	5.93	4.7	0	99.53
30/4/2009	70	10	483.04	254.10	6.9	1752	6.8	2.51	6.04	4.9	0	99.48
1/5/2009	71	10	555.77	254.10	6.7	1318	7.0	1.41	4.06	5.3	0	99.75
4/5/2009	74	10	533.02	272.20	7.0	1668	7.2	0.65	7.08	5.6	0	99.88
5/5/2009	75	10	493.95	273.90	7.3	1420	6.9	1.57	4.88	5.5	0	99.68
6/5/2009	76	10	503.26	288.50	7.1	1622	6.7	2.27	5.28	4.9	0	99.55

Table B-7 Effects of solid retention time on MBR performance

Time (hours)	FM (mg/L)						TOC (mgC/L)					
	SRT 60 days		SRT 30 days		SRT 10 days		SRT 60 days		SRT 30 days		SRT 10 days	
	Average	SD	Average	SD	Average	SD	Average	SD	Average	SD	Average	SD
0	438.96	35.09	430.73	9.99	418.78	5.62	310.27	8	299.30	17	287.33	3
0.25	145.33	21.88	199.71	22.11	229.35	11.24	144.00	24	167.70	8	186.30	24
0.5	49.54	20.54	113.90	7.33	123.60	28.07	126.83	19	150.93	2	176.00	23
1	3.67	2.12	28.86	24.04	69.44	23.88	106.73	22	118.30	15	152.20	25
2	1.58	0.92	5.92	4.52	20.43	8.78	82.98	21	76.20	12	110.11	16
3	1.26	0.76	2.26	0.36	7.02	3.54	55.18	9	37.15	10	77.01	18
5	0.99	0.66	1.71	0.39	1.43	0.47	11.74	10	8.98	1	12.94	3
8	0.68	0.41	1.60	0.36	1.13	0.39	5.66	2	7.11	1	9.48	3

Table B-8 Specific flux during membrane operation

Date	Day	SRT (days)	Flow rate		TMP		Flux			Remark
			(L/hr)	SD	(in Hg)	(kPa)	LMH	LMH/kPa	% Recovery	
26/2/2009	1	60	66.67	0.00	1.80	6.10	78.43	12.867	-	Replacement
26/2/2009	1	60	66.05	7.91	4.80	16.25	77.71	4.781	-	
28/2/2009	3	60	37.63	15.23	6.50	22.01	44.27	2.011	-	
2/3/2009	5	60	24.13	2.16	4.53	15.35	28.39	1.849	-	
3/3/2009	6	60	23.08	0.00	3.80	12.87	27.15	2.110	-	
4/3/2009	7	60	63.16	0.00	2.20	7.45	74.30	9.974	77.5	cleaning
4/3/2009	7	60	26.07	6.20	21.87	74.05	30.67	0.414	-	
6/3/2009	9	60	3.11	0.22	1.60	5.42	3.66	0.675	-	
9/3/2009	12	60	62.72	0.38	3.80	12.87	73.79	5.734	44.6	cleaning
9/3/2009	12	60	26.07	6.20	21.60	73.15	30.67	0.419	-	
10/3/2009	13	60	37.63	15.23	5.00	16.93	44.27	2.614	-	
11/3/2009	14	60	10.96	1.82	10.80	36.57	12.90	0.353	-	
12/3/2009	15	60	10.26	0.75	15.20	51.47	12.07	0.235	-	
13/3/2009	16	60	8.49	0.14	15.60	52.83	9.99	0.189	-	
16/3/2009	19	60	6.01	0.71	16.60	56.21	7.08	0.126	-	
17/3/2009	20	60	62.94	0.77	5.80	19.64	74.05	3.770	29.3	cleaning
18/3/2009	21	60	16.98	0.27	9.80	33.19	19.98	0.602	-	
19/3/2009	22	60	10.26	0.75	10.80	36.57	12.07	0.330	-	
21/3/2009	24	60	3.74	0.00	11.20	37.93	4.40	0.116	-	
23/3/2009	26	60	61.65	0.74	9.20	31.15	72.53	2.328	18.1	cleaning
23/3/2009	26	60	32.50	4.33	13.20	44.70	38.24	0.855	-	
24/3/2009	27	60	0.82	0.03	9.80	33.19	0.97	0.029	-	
26/3/2009	29	60	0.92	0.04	20.20	68.41	1.09	0.016	-	
27/3/2009	30	30	62.50	0.00	1.80	6.10	73.53	12.063	-	Replacement
27/3/2009	30	30	20.75	2.48	4.60	15.58	24.41	1.567	-	
28/3/2009	31	30	14.76	0.41	7.40	25.06	17.37	0.693	-	
30/3/2009	33	30	12.77	1.39	11.20	37.93	15.03	0.396	-	

Date	Day	SRT (days)	Flow rate		TMP		Flux			Remark
			(L/hr)	SD	(in Hg)	(kPa)	LMH	LMH/kPa	% Recovery	
31/3/2009	34	30	12.77	1.39	16.20	54.86	15.03	0.274	-	
1/4/2009	35	30	15.14	0.93	19.80	67.05	17.81	0.266	-	
2/4/2009	36	30	3.67	0.17	20.20	68.41	4.32	0.063	-	
3/4/2009	37	30	62.29	0.37	6.20	21.00	73.28	3.490	28.9	cleaning
3/4/2009	37	30	27.84	3.74	7.13	24.16	32.76	1.356	-	
6/4/2009	40	30	25.26	4.31	9.20	31.15	29.72	0.954	-	
7/4/2009	41	30	24.16	1.09	13.20	44.70	28.43	0.636	-	
9/4/2009	43	30	27.51	2.50	21.07	71.34	32.36	0.454	-	
10/4/2009	44	30	63.83	0.00	7.20	24.38	75.09	3.080	25.5	cleaning
10/4/2009	44	30	23.35	2.33	9.07	30.70	27.47	0.895	-	
20/4/2009	54	30	26.28	1.29	13.20	44.70	30.92	0.692	-	
21/4/2009	55	30	61.43	0.36	6.20	21.00	72.28	3.442	28.5	cleaning
21/4/2009	55	30	48.33	2.36	9.53	32.28	56.86	1.761	-	
22/4/2009	56	30	3.86	0.32	11.07	37.48	4.54	0.121	-	
23/4/2009	57	30	2.83	0.14	12.20	41.31	3.34	0.081	-	
24/4/2009	58	10	55.79	8.37	1.80	6.10	65.64	10.768	-	Replacement
24/4/2009	58	10	43.48	0.00	11.20	37.93	51.15	1.349	-	
27/4/2009	61	10	15.67	4.01	14.80	50.12	18.43	0.368	-	
28/4/2009	62	10	11.74	0.83	15.20	51.47	13.82	0.268	-	
29/4/2009	63	10	11.59	0.94	15.80	53.50	13.63	0.255	-	
30/4/2009	64	10	9.11	1.16	16.20	54.86	10.72	0.195	-	
1/5/2009	65	10	9.74	0.66	16.80	56.89	11.46	0.201	-	
4/5/2009	68	10	64.75	0.40	6.80	23.03	76.18	3.308	30.7	cleaning
4/5/2009	68	10	31.83	2.66	10.20	34.54	37.45	1.084	-	
5/5/2009	69	10	8.29	0.35	15.80	53.50	9.75	0.182	-	
6/5/2009	70	10	7.72	0.22	16.20	54.86	9.09	0.166	-	

Table B-9 Effect of solid retention time of formaldehyde removal on SBR and MBR performance

SRT (days)	SBR				MBR			
	FM (mg/L)		% Removal		FM (mg/L)		% Removal	
	Average	SD	Average	SD	Average	SD	Average	SD
60	0.73	0.46	1.09	0.69	99.79	0.13	99.79	0.13
30	1.62	0.32	1.34	0.46	99.75	0.09	99.75	0.09
10	1.48	0.94	1.93	0.75	99.62	0.15	99.62	0.15

APPENDIX C

Statistical analysis

Example of Statistical Analysis by ANOVA

Question: Is there any difference in the means of FM removal efficiency between SRT 60 days and SRT 30 days of SBR operation?

Hypotheses :

H_0 : There is no difference in the means of FM removal effeiciency in the groups tested.

H_1 : There is difference in the means of FM removal effeiciency in the groups tested.

Significant value:

P-value (p) = *0.05*

Input data for ANOVA in SPSS

Group 1: SRT 60 days: 99.95, 99.80, 99.95, 99.94, 99.85, 99.91, 99.84, 99.79, 99.56, 99.78, 99.78, and 99.80

Group 2: SRT 30 days: 99.72, 99.51, 99.60, 99.64, 99.70, and 99.53

Results from ANOVA

		Sum of Squares	df	Mean Square	F	Sig	*P*	Result
SRT 60 vs 30	Between Groups	0.181	1	0.181	17.365	0.010	0.05	D
	Within Groups	0.166	16	0.010				
	Total	0.347	17					

Intepretation

From the ANOVA result

if sig < *P*-value (p=0.05) : H_0 is rejected, H_1 is accepted (Difference = D)

if sig > *P*-value (p=0.05) : H_0 is accepted, H_1 is rejected (No difference = ND)

According to Table C-1, sig. equals to 0.010 , which is less than 0.05. The ANOVA result indicated that the mean of FM removal efficiency at SRT of 60 days was significant different with that at SRT of 30 days with 95% confidence.

Table C-1 Results from analysis of variance (ANOVA) assess the effect of SRT on SBR performance

		Sum of Squares	df	Mean Square	F	Sig	*P*	Result
SRT 60 vs 10	Between Groups	0.163	1	0.163	5.814	0.027	0.05	D
	Within Groups	0.477	17	0.028				
	Total	0.640	18					
SRT 60 vs 30	Between Groups	0.181	1	0.181	17.365	0.010	0.05	D
	Within Groups	0.166	16	0.010				
	Total	0.347	17					
SRT30 vs 10	Between Groups	0.001	1	0.001	0.039	0.848	0.05	ND
	Within Groups	0.385	11	0.035				
	Total	0.386	12					

Table C-2 Results from analysis of variance (ANOVA) assess the effect of SRT on MBR performance

		Sum of Squares	df	Mean Square	F	Sig	*P*	Result
SRT 60 vs 10	Between Groups	0.123	1	0.123	6.453	0.021	0.05	D
	Within Groups	0.324	17	0.019				
	Total	0.447	18					
SRT 60 vs 30	Between Groups	0.008	1	0.008	0.574	0.046	0.05	ND
	Within Groups	0.234	16	0.015				
	Total	0.243	17					
SRT 30 vs 10	Between Groups	0.047	1	0.047	2.966	0.113	0.05	ND
	Within Groups	0.175	11	0.016				
	Total	0.223	12					

Table C-3 Results from analysis of variance (ANOVA) assess the comparison between SBR and MBR

SRT (days)			Sum of Squares	df	Mean Square	F	Sig	*P*	Result
60	SBR vs. MBR	Between Groups	0.008	1	0.008	0.554	0.465	0.05	ND
		Within Groups	0.321	22	0.015				
		Total	0.329	23					
30	SBR vs. MBR	Between Groups	0.051	1	0.051	0.633	0.310	0.05	ND
		Within Groups	0.080	10	0.008				
		Total	0.131	11					
10	SBR vs. MBR	Between Groups	0.000	1	0.000	0.011	0.917	0.05	ND
		Within Groups	0.480	12	0.040				
		Total	0.481	13					

BIOGRAPHY

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