REMOVAL OF FORMALDEHYDE SYNTHETIC WASTEWATER BY CHITOSAN-ENHANCED MEMBRANE FILTRATION

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หัวข้อวิทยานิพนธ์	การกำจัดฟอร์มัลดีไฮด์ในน้ำเสียสังเคราะห์ด้วยกระบวนการเมมเบรน โดยใช้ไคโตซานเป็นตัวเพิ่มประสิทธิภาพ
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บทคัดย่อ

งานวิจัยนี้เป็นการศึกษาการกำจัดฟอร์มัลดีไฮด์ในน้ำเสียสังเคราะห์ด้วยกระบวนการเมมเบรนโดยใช้ไคโตซานเป็นตัวเพิ่มประสิทธิภาพ การทดลองแบ่งออกเป็น 2 ตอน คือ 1) การทดลองการดูดซับและ 2) การทดสอบประสิทธิภาพของระบบเมมเบรนเพื่อศึกษาปัจจัยที่มีผลต่อความสามารถในการกักกันฟอร์มัลดีไฮด์, ปริมาณคาร์บอนอินทรีย์ละลายน้ำ, ความขุ่นและอัตราการลดลงของฟลักซ์ จากผลการทดลองการดูดซับฟอร์มัลดีไฮด์ด้วยไคโตซานพบว่าอัตราส่วนของไคโตซานต่อฟอร์มัลดีไฮด์ที่เพิ่มขึ้นส่งผลให้ความสามารถในการดูดซับเพิ่มขึ้นแต่ก็มีผลทำให้ปริมาณคาร์บอนอินทรีย์ละลายน้ำเพิ่มขึ้นด้วย สำหรับขนาดของเกล็ดไคโตซาน พีเอชของสารละลายฟอร์มัลดีไฮด์และที่ระยะเวลาในการดูดซับมากกว่า 0.5 ชั่วโมงขึ้นไปไม่มีผลต่อความสามารถในการดูดซับฟอร์มัลดีไฮด์ จากการทดลองการดูดซับพบว่าที่อัตราส่วนไคโตซานต่อฟอร์มัลดีไฮด์เท่ากับ 500:1 มีความสามารถในการกำจัดฟอร์มัลดีไฮด์ร้อยละ 71-72 ที่ระยะเวลาในการดูดซับ 0.5 ชั่วโมง โดยใช้ไคโตซานที่ไม่ทำการคัดแยกขนาดและไม่ทำการปรับค่าพีเอชของสารละลายฟอร์มัลดีไฮด์ จากการทดสอบประสิทธิภาพของระบบเมมเบรนพบว่าความสามารถในการกักกันฟอร์มัลดีไฮด์มีค่าเพิ่มขึ้นเมื่ออัตราส่วนของไคโตซานต่อฟอร์มัลดีไฮด์เพิ่มขึ้นโดยมีความสามารถในการกำจัดฟอร์มัลดีไฮด์ร้อยละ 35-36, 50-51 และ 59-60 และมีอัตราการลดลงของฟลักซ์ร้อยละ 10-18, 83-90 และ 91-99 ที่อัตราส่วนไคโตซานต่อฟอร์มัลดีไฮด์ 100:1, 200:1 และ 300:1 ตามลำดับ สำหรับขนาดของเกล็ดไคโตซานและพีเอชของสารละลายฟอร์มัลดีไฮด์ไม่มีผลต่อความสามารถในการกักกันฟอร์มัลดีไฮด์และอัตราการลดลงของฟลักซ์ของทั้งไมโครและอัลตร้าฟิวเตรชันเมมเบรน

คำสำคัญ: การกักกัน, การดูดซับ, ไคโตซาน, ฟอร์มัลดีไฮด์, เมมเบรน

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ABSTRACT

In this research, chitosan was used to enhance the membrane performance in retention of formaldehyde-absorbed chitosan. The experiments were being conducted into 2 steps: 1) Adsorption test to study formaldehyde adsorption by chitosan and 2) Membrane performance test to investigate factors affecting formaldehyde rejection, dissolved organic carbon (DOC), turbidity, and flux decline. The results from adsorption test showed that formaldehyde adsorption efficiency and DOC increased when chitosan:formaldehyde ratio increased, whereas chitosan size, pH of formaldehyde solution, and mixing time more than 0.5 hours did not exhibit any effect on formaldehyde adsorption. However, increasing chitosan:formaldehyde ratio and smaller size of chitosan flake caused higher turbidity and DOC levels in treated water. With a 500:1 weight ratio of chitosan to formaldehyde at mixing time 0.5 hours, it was found that the mixed size of chitosan flake was possible to remove formaldehyde up to 71-72%. In membrane process, formaldehyde retention and percent flux decline increased when chitosan:formaldehyde ratio increased with percent removal of 35-36%, 50-51%, and 59-60%. While percent flux decline were in range of 10-18%, 83-90%, and 91-99% at chitosan:formaldehyde ratio of 100:1, 200:1, and 300:1, respectively. Chitosan size and pH of formaldehyde solution did not exhibit any effect on formaldehyde retention and flux decline of both membranes.

KEYWORDS: Adsorption, Chitosan, Formaldehyde, Membrane, Retention

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Pawarrit Patthrasittisorn

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CHAPTER 1

INTRODUCTION

1.1 Motivation

Formaldehyde is a compound that commonly used in large variety of processes in chemical industry, such as synthetic resin production, aquaculture industry, adhesive and glue manufacturing, disinfectant and preservative production, chemical and petrochemical industry, textile processing, paper manufacturing, and wood processing[1]. Those industries produce wastewater with high formaldehyde concentration for example, during the production of polyester fiber (3-5 g formaldehyde/L)[2], industrial adhesives (0.2-40 g formaldehyde/L)[2]. According to its toxicity on living organisms and environment, the national industrial effluent standard states that effluent contaminated formaldehyde of more than 1 mg/L is not allowed to discharge into receiving water. Therefore, appropriate treatment methods should be applied to remove formaldehyde in wastewater before discharging into the natural water. A number of methods have been used for treatment of formaldehyde in wastewater such as adsorption by activated carbon[2], oxidation using ozone (O_3), hydrogen peroxide (H_2O_2) or ultraviolet coupled with hydrogen peroxide (UV/H_2O_2)[3] and treatment with biological processes using activated sludge[4]. These processes are found to remove formaldehyde efficiently, but they have some limitations regarding to operation and maintenance, as well as the occurrence of some residuals of toxic substances.

Membrane technology is one of the most effective methods for separation and fractionation solutes from solution such as suspended particles, monovalent and multivalent ions, or each bacteria and viruses. Membrane is a thin layer of materials (such as polymer, metal, or ceramic) which separation mechanisms are depending on their chemical or physical properties, i.e. size exclusion or charge interaction. Membrane can be categorized by pore sizes or molecular weight cut-off (MWCO) from large to small as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). This technology has considerable attention for wastewater treatment and water reclamation because of simplicity of operation, and high quality effluent. MF and UF membranes typically have pore sizes in a range of 0.1-10 μm and 0.01-0.1 μm or MWCO of 20,000-100,000 and 100,000-500,000 daltons, respectively. However, these membranes are not suitable to separate molecules smaller than their MWCO because they are porous membranes. Formaldehyde can easily pass through the pores of MF and UF membranes because it has molecular size less than pore size of the membranes. On the other hand, formaldehyde molecules are not retained by MF and UF membranes. An application recently on MF and UF membranes for treatment of formaldehyde wastewater is in the presence of water-soluble polymer, referred as polymer-enhanced membrane filtration. Since MF or UF membranes are not suitable for separation of formaldehyde, therefore water-soluble polymers are used to bind formaldehyde and form macromolecules, leading to retention by MF or UF membranes[5]. In treatment of formaldehyde containing wastewater, chitosan is used as a binding polymer to form macromolecules. Chitosan is a partially deacetylated polymer obtained from the alkaline deacetylation of chitin or extracted from shellfish sources[6]. The deacetylated process generates amino groups which are capable to react with environmental contaminants, i.e. formaldehyde[2].

This research studied the feasibility of formaldehyde removal by chitosan-enhanced MF and UF membrane filtration as a post-treatment method for better quality effluent. Factors affecting formaldehyde adsorption by chitosan, i.e. chitosan:formaldehyde ratio, size of chitosan flake, pH of formaldehyde solution, and mixing time were investigated. Membrane process, consisting of MF and UF membranes was operated with conditions achieved from adsorption tests. Parameters including formaldehyde removal, flux decline, total organic carbon, and turbidity removal were investigated to indicate membrane performance. Factors affecting membrane performance i.e. chitosan:formaldehyde ratio, size of chitosan flake, and pH of formaldehyde solution were investigated. The results of this research exhibited a potential of chitosan-enhanced membrane filtration process in removal of formaldehyde for industrial wastewater.

1.2 Objectives

1.2.1 To study factors affecting formaldehyde adsorption by chitosan i.e. chitosan:formaldehyde ratio, size of chitosan flake, pH of formaldehyde solution, and mixing time.

1.2.2 To study feasibility in removal of formaldehyde from synthetic wastewater by chitosan-enhanced MF and UF membranes.

1.2.3 To study factors affecting membrane performance during chitosan-enhanced membrane filtration i.e. chitosan:formaldehyde ratio, size of chitosan flake, and pH of formaldehyde solution.

1.3 Scope of study

1.3.1 Formaldehyde wastewater used in the experiment was synthetic wastewater, diluted from 40% (w/v) formaldehyde solution.

1.3.2 Chitosan used in the experiment was in a form of chitosan flake with 85% deacetylation and molecular weight of 50,000 daltons.

1.3.3 Microfiltration (MF) and ultrafiltration (UF) membranes were used in the experiment. Both MF and UF membranes were made of polypropylene (PP). MF was conducted in cartridge module with pore size of 5 μm, whereas UF was operated in hollow fiber module with pore size of 0.01 μm.

1.3.4 Factors affecting formaldehyde adsorption by chitosan were studied as follows:

1) Chitosan:formaldehyde ratio (by weight): 100:1, 200:1, 300:1, 400:1 and 500:1.

2) Size of chitosan flake: ≤ 0.85 mm, 0.85-1.7 mm, 1.7-4.0 mm, ≥ 4.0 mm and mixed size.

3) pH of formaldehyde solution: Original pH, pH 4, pH 7 and pH 10.

4) Mixing time: 0.25, 0.5, 0.75, 1, 2, 4, 8 and 12 hrs.

1.3.5 Membrane performance was investigated in terms of formaldehyde removal, flux decline, and turbidity.

1.3.6 Factors affecting membrane performance were studied as follows:

1) Types of membrane: Microfiltration (MF) and Ultrafiltration (UF).

2) Chitosan:formaldehyde ratio (by weight): 100:1, 200:1, 300:1, 400:1 and 500:1.

3) Size of chitosan flake: ≤ 0.85 mm, 0.85-1.7 mm, 1.7-4.0 mm, ≥ 4.0 mm and mixed size.

4) pH of formaldehyde solution: original pH, pH 4, pH 7 and pH 10.

1.4 Expected results

1.4.1 Understanding of feasibility in formaldehyde removal by chitosan adsorption and chitosan-enhanced membrane filtration.

1.4.2 Application of chitosan-enhanced membrane filtration process in removal of formaldehyde and other environmental contaminants from industrial wastewater.

CHAPTER 2

THEORY AND LITERATURE REVIEWS

2.1 Formaldehyde

Formaldehyde is a compound that commonly used in large variety of processes in chemical industry, such as synthetic resin production, aquaculture industry, adhesive and glue manufacturing, disinfectant and preservative production, chemical and petrochemical industry, textile processing, paper manufacturing and wood processing[1]. Most formaldehyde is used in the production of polymers and other chemicals. Reactions of formaldehyde with phenol, urea, or melamine result as hard thermoset resins. The resins are commonly used in permanent adhesives for plywood or carpeting. They are also foamed to make insulation, or casted into molded products. Wastewater from those industries contains high concentration of formaldehyde for example, during the production of industrial adhesives (0.2-40 g formaldehyde/L)[], chemical industries (5-10 g formaldehyde/L)[2].

Formaldehyde is produced by catalytic oxidation of methanol. The most common catalysts are silver metal or a mixture of iron and molybdenum or vanadium oxides. The silver-based catalyst is usually operated at temperature of 650°C. A chemical reaction to produce formaldehyde by catalytic oxidation of methanol is shown in Eq.2.1.

$$CH_3OH \rightarrow HCHO + H_2 \qquad (2.1)$$

In iron oxide system, methanol and oxygen react at 400°C to produce formaldehyde according to the reaction shown in Eq.2.2.

$$CH_3OH + \frac{1}{2}O_2 \xrightarrow{Fe_2O_3} HCHO + H_2O \qquad (2.2)$$

Formaldehyde reacts with water to form an equilibrium mixture of water, formaldehyde and formaldehyde hydrate as shown in Eq. 2.3.

$$\underset{\text{Formaldehyde}}{CH_2O_{(g)}} + H_2O_{(l)} \leftrightarrow \underset{\text{Formaldehyde hydrate}}{CH_2(OH)_{2\,(l)}} \qquad (2.3)$$

Physical and chemical properties of formaldehyde are listed in Table 2.1.

Table2.1 Chemical and physical properties of formaldehyde[7]

Properties	Characteristic
Chemical Name	Formaldehyde
Synonym (s)	Formic aldehyde, Methanal, Methyl aldehyde, Methylene oxide
Register trade name (s) for 37% aqueous solution	Formalin, Formol, Morbicid, Veracur
Register trade name (s) for polymeric form	Paraformaldehyde, Polyoxymethylene, Paraform, Formagene
Chemical formula	CH_2O
Molecular weight	30.03
Color	Colorless
pH (37% solution, 25°C)	2.8-4.0
Boiling point	98 °C
Melting point	-15 °C
Log K_{ow}	0.350
Log K_{oc}	1.567
Henry's law constant	3.27×10^{-7} atm.m^3/mol
Solubility (water, 20°C)	More than 100 mg/ 100 ml
Vapor pressure (37°C)	55 mmHg
Vapor density (Air = 1)	1
Specific gravity (water = 1)	1.08

The 37-50% solution of formaldehyde in water with drops of methyl alcohol is known as formalin. Methyl alcohol was added to the solution to prevent polymerization of formaldehyde. The polymeric form of formaldehyde with degree of polymerization of 8-100 units is called paraformaldehyde (para-formaldehyde, *p*-formaldehyde) and usually obtained as white powder. When paraformaldehyde was heated or dissolved in water, it depolymerises back to formaldehyde. Paraformaldehyde is also known as polyoxymethylene. It was used as thermoplastic (POM, Delrin, polyformaldehyde). Figure 2.1 shows the structures of formaldehyde and paraformaldehyde.

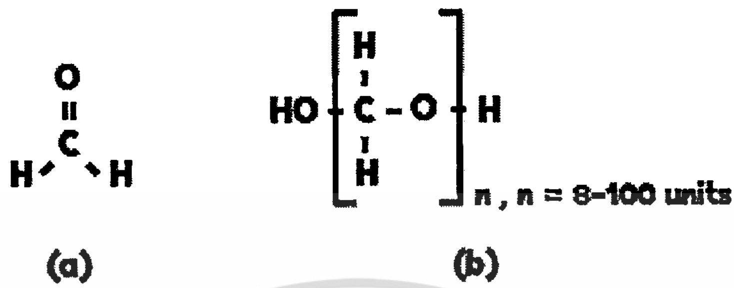


Figure 2.1 Molecular structures of (a) formaldehyde and (b) paraformaldehyde[7]

2.2 Membrane technology[8-9]

2.2.1 Membrane description[8-9]

Membrane is a thin layer of material which is capable of separating materials as a function of their chemical or physical properties when a suitable driving force is applied. Membranes are available in four configurations; plate and frame, tubular, hollow fiber, and spiral wound. They are made of various materials such as polymer, metal, and ceramic. The basic concept of membrane separation is shown in Fig. 2.2.

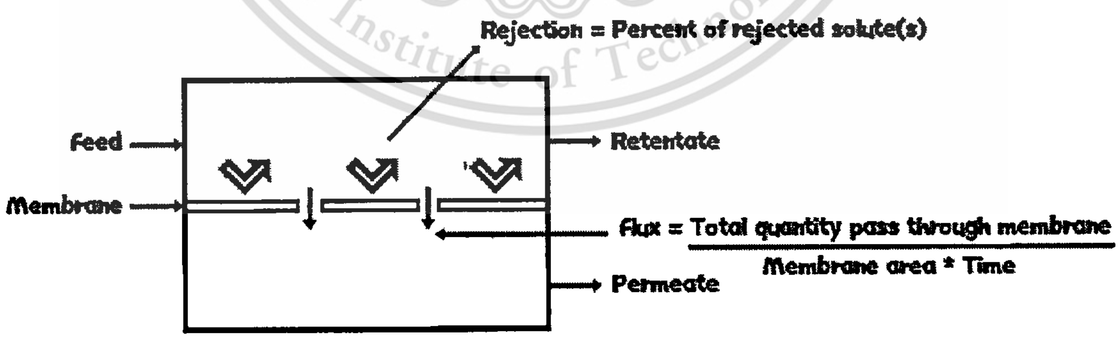


Figure 2.2 Transport of solutes in membrane system[10]

Feed stream enters the membrane system and a suitable driving force (such as concentration or pressure gradient) is applied to the membrane, this leads to preferential transport of one or more components. Certain components (solutes, solvents, or gases) pass through the membrane. Other components do not pass through the membrane or pass through very slowly. The selective transport (called permeation) forms the basis of membrane separations, which generally involve the separation of solutes or fluid. The stream containing the components that permeate through membrane is called permeate (or filtrate), whereas the stream containing retained components is called retentate (or concentrate).

2.2.2 Operation of membrane[11]

Membrane process has three possible streams: feed, permeate and retentate. Retentate is defined as unpermeated product. If there is no retentate, the operation is termed dead-end filtration as shown in Fig. 2.3 (a). Feed solution flows in tangential with membrane surface and pressure was applied in the same direction, permeate was passed through the membrane. As particles accumulate on the membrane surface, and the pressure required to maintain the required permeate flow increases, the membrane must be replaced.

In case significant solids contain on membranes have limited permeability (dense membranes), cross-flow filtration was operated. As shown in Fig. 2.3 (b), feed solution flows in parallel direction of membrane surface or tangential with applied pressure, permeate was passed through the membrane while some of the feed solution is collected as a retentate. Because the feed was applied in parallel direction, it removes the accumulated particles on membrane surface by scouring of the retentate flowing over the surface.

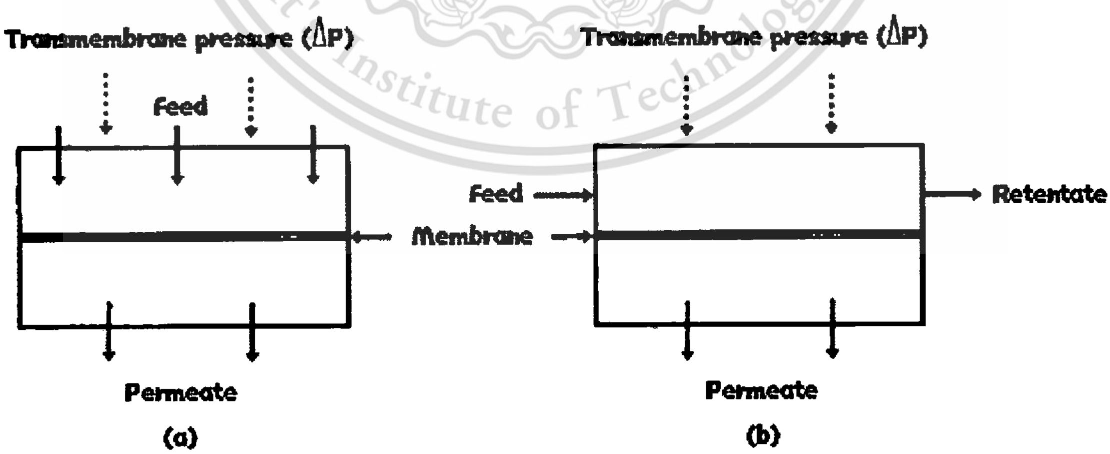


Figure 2.3 Modes of membrane operation (a) dead-end filtration (b) cross-flow filtration[16]

2.2.3 Membrane module

Membranes are available in four major types; plate and frame, tubular, hollow fiber, and spiral wound.

1. Plate and frame

Plate and frame module consist of membrane, spacers, and supporter that layered together between two end plates. The specific surface area (membrane surface per module volume) of this units is about 100-400 m^2/m^3. Each plate has thickness in a range of 0.5 to 3.0 mm. Plate and frame module is possible arranged in parallel and/or in series circulation. Feed solution is directed across each plate in series, permeate enters the membrane and is collected through the central permeate collection channel. The configuration of plate and frame module is shown in Fig. 2.4.

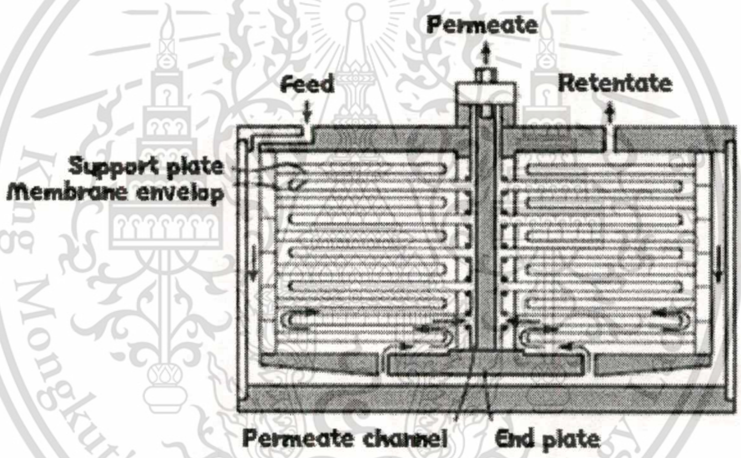


Figure 2.4 Plate and frame module[12]

2. Tubular

Tubular module is not self-supporting membrane. It consists of several membrane tubes (5-15 mm in diameter) located inside a housing. The specific surface area of this membrane is about 150-300 m^2/m^3. Feed stream enters inside the tube, permeate passes through the side of membrane tube and is collected together as shown in Fig. 2.5.

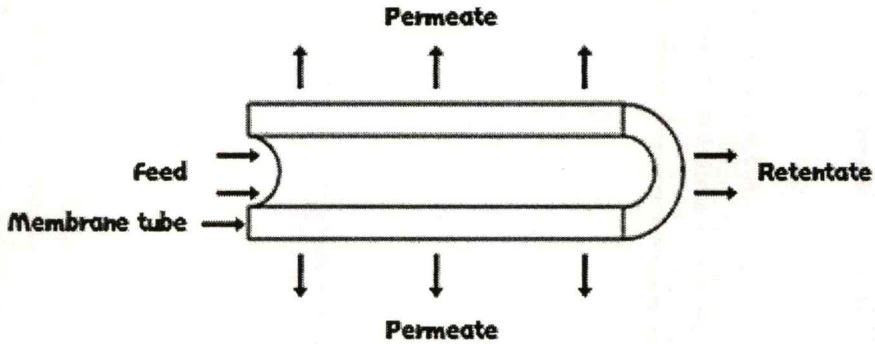


Figure 2.5 Tubular module[13]

3. Hollow fiber

Hollow-fiber module is a self-supporting membrane, consisting of several small diameter membrane fiber bundles. The specific surface area of this membrane is about 9000-30,000 m^2/m^3. Feed solution can enter inside the fibers (outside-in configuration) or on the outside (inside-out configuration) based on the direction of filtration flow. The outside-in configuration is shown in Fig. 2.6 (a). The fiber bundles are often arranged in a U shape and sealed at only one end. The pressurized feed stream flows from the outside of a hollow fiber, permeate is passed through and collected inside of the hollow fibers. For the inside-out configuration, pressurized feed water flows through the pores of hollow fibers, permeate is passed through and collected outside of the membrane fibers as shown in Fig. 2.6 (b).

Figure 2.6 Hollow fiber modules (a) outside-in configuration and (b) inside-out configuration[14]

4. Spiral wound

Spiral wound module uses flat sheet membranes, supporter, and spacer wrapped spirally around a collection tube as shown in Fig. 2.7. Specific surface area of this module is 300-1000 m^2/m^3. Feed solution passes axially down the module across the membrane, a portion of the feed solution permeates through membrane where it spirals toward the center and exits through permeate collection tube.

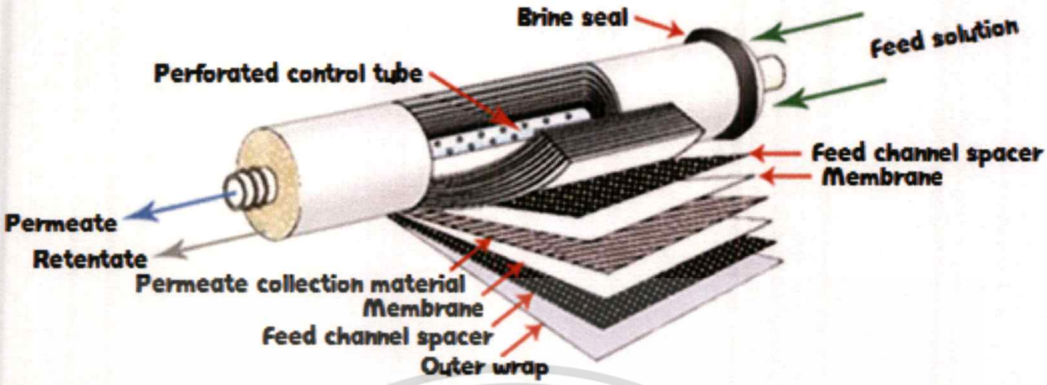


Figure 2.7 Spiral wound module[15]

Comparison of membrane modules between plate and frame, tubular, hollow fiber and spiral wound membranes are listed in Table 2.2.

Table2.2 Comparison between several membrane modules[16]

Parameter	Plate and frame	Tubular	Hollow fiber	Spiral wound
Specific surface area (m^2/m^3)	100-400	150-300	9,000-30,000	300-1,000
Flux ($L/m^2{\cdot}h$)	10-50	10-50	0.5-5.0	10-50
Fouling tendency	Moderate	Low	High	Moderate
Membrane cleaning	Moderate	Easy	Difficult	Moderate
Membrane cost	Moderate	High	Low	Low

2.2.4 Application of membrane

Membrane technology has become a dignified separation technology over the past decennia and has been used increasingly in a broad range of applications. The main force of membrane technology is the fact that it works without addition of chemicals, with a relatively low energy use, as well as easy and well-arranged process conductions. The important property of membrane, which is exploited in every application, is the ability of a membrane to control the permeation of chemical species in contact with it. Applications of membrane are shown in Table 2.3.

Table2.3 Applications of membrane separation processes[17]

Microfiltration :	Sterilization of drug
	Purification of antibiotic
	Separation of mammalian cell from liquid
Ultrafiltration :	Preconcentration of milk before making cheese
	Recovery of vaccine and antibiotic from fermentation broth
	Color removal from kraft black in paper making
	Fractionation of proteins
	Clarification of fruit juice
Nanofiltration :	Organic solvent separation
	Desalination of brackish and seawater
	Treatment of wastewater to remove a wide variety of impurities
Reverse Osmosis :	Organic solvent separation
	Desalination of brackish and seawater
	Treatment of wastewater to remove a wide variety of impurities
	Ultrapure water production
	Concentration of foodstuff
	Removal of alcohol from beer and wine

2.2.5 Membrane fouling[11]

In membrane process, one of problems on membrane operation is membrane fouling. The major symptoms of membrane fouling are flux decline over operating time. A reduction in permeate flux is possible caused by accumulation of reign particles on membrane surface or within the membrane pores, or it may cause changes in chemical characters of the membrane material. The substances that have potential to foul membrane can be categorized as:

1. Molecules or particles
2. Organic compounds
3. Inorganic compounds
4. Biota that grow on the membrane surface

Molecules or particles foul membrane 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. Besides, the substances that fouled membrane, the nature and extent of fouling is influenced by chemical and biological characters of solution, chemical composition of the membrane, solute-solute interactions, and membrane-solute interactions.

2.2.5.1 Fouling mechanisms

1. Size exclusion

Size exclusion is a fouling mechanism for dense membranes i.e. NF and RO membranes. When the membrane is used to separate molecules or particles larger than pore size of the membrane, these solutes will accumulate on the surface or in membrane pores, leading to high membrane resistance and reduction of permeate flux.

2. Charge interaction

Charge interaction is a reaction between charges of solutes and membrane surface. Material used in membrane production promotes charges on membrane surface. In case that membrane surface employed the same charge with solutes, an electrostatic repulsion between solutes and surface occurs. On the other hand, if charges of solutes and membrane surface are different, it causes membrane fouling by electrostatic attraction.

3. Hydrophobic interaction

Hydrophobic interaction is a reaction of polar and non-polar molecules. When non-polar molecules are surrounded by polar molecules, it has repulsion between polar and non-polar molecules. Non-polar molecules are assembling and separate from polar molecules.

2.2.5.2 Fouling characteristics

1. Concentration polarization

Concentration polarization (CP) is a phenomenon when rejected molecules or particles accumulate in boundary layer of membrane, as shown in Fig. 2.8. The CP causes concentration of solutes on membrane surface (C_w) higher than in bulk solution (C_b). The concentration polarization layer increases resistance of membrane and reduces the permeate flux.

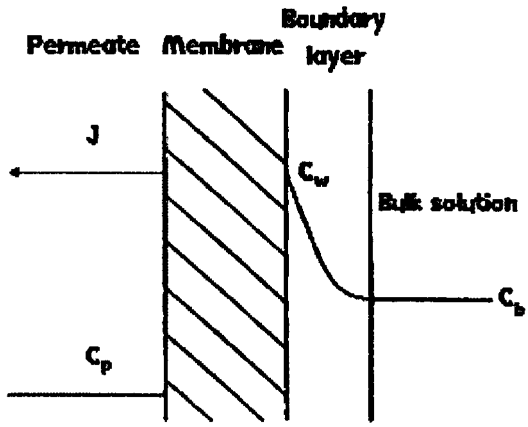


Figure 2.8 Characteristic of concentration polarization[16]

2. Gel layer

Gel layer is the layer of large molecules or particles accumulated on membrane surface. When concentration of molecules or particles on surface reaches to limit of dissolution (gel concentration, C_g) gel layer is formed. Figure 2.9 shows characteristic of gel layer on membrane surface. Gel layer causes high membrane resistance and decrease in solution flux, also it causes a change of membrane recovery.

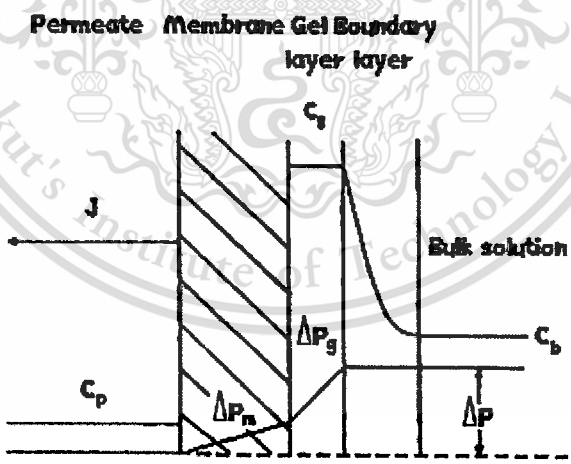


Figure 2.9 Characteristic of gel layer[16]

3. Cake formation

Cake formation is similar to gel layer but the difference is size of molecules or particles accumulating on membrane surface. Gel layer is caused by an accumulating of large molecules or particles, whereas cake formation is caused by an accumulating of suspended particles. If solution is a mixture between small and large particles, small particles will accumulate in membrane pores while large particles deposite on membrane surface afterward. Gel layer has thickness in a range of micrometer but cake layer has thickness in a range of millimeter.

4. Pore blockage

Pore blockage of membrane causes membrane fouling in many characters. If pore sizes of membrane are smaller than particles, the particles will accumulate on membrane surface as shown in Fig. 2.10 (a). Figure 2.10 (b) shows pore blockage of membrane when pore sizes are slightly bigger than particles. In case of membrane pore sizes are larger than particles, these particles will firstly accumulate within membrane pores and then deposite on membrane surface afterwards, as shown in Fig. 2.10 (c).

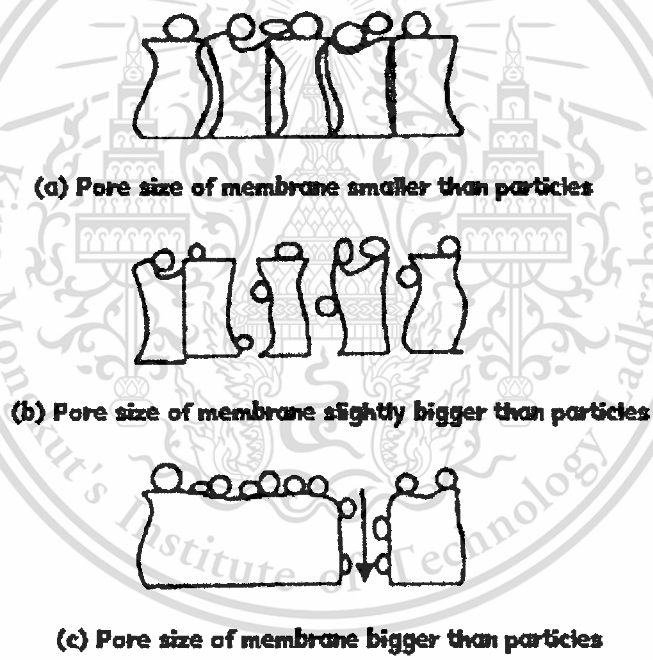
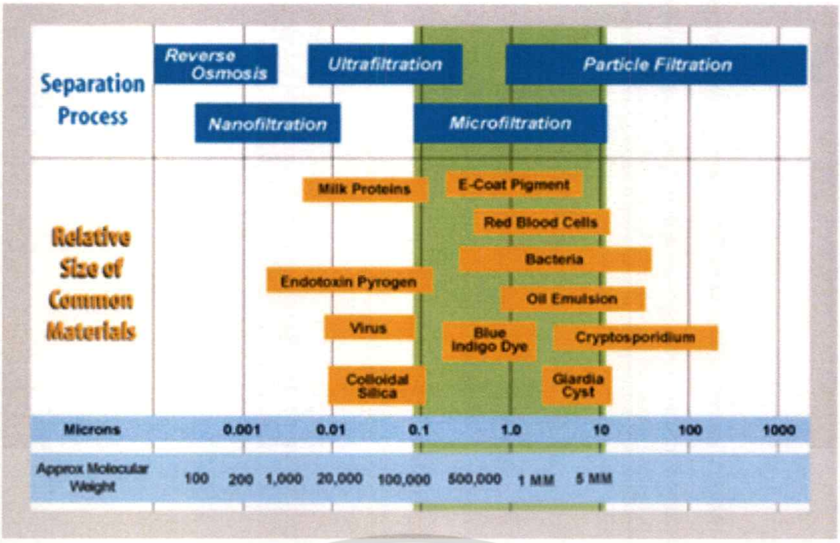


Figure 2.10 Characteristic of membrane pore blockage[16]

2.2.6 Microfiltration

Microfiltration (MF) is a low pressure process (100-500 kPa or 1-5 atm) for separating colloid, suspended particles and macromolecules with diameters in a range of 0.1-10 μm (or molecular weight cut-off of 100,000 - 500,000 daltons) by macroporous membranes. Figure 2.11 shows the schematic of microfiltration characteristic.



Figures 2.11 Schematic diagram of retained particles size and MWCO on microfiltration[18]

Flux of solution for MF membrane can be calculated from Darcy's law as shown in Eq.2.4.

$$J_V = (\frac{1}{A}) \cdot (\frac{dV}{dt}) \tag{2.4}$$

where J_v is flux of solution ($m^3/m^2 \cdot h$), A is membrane area (m^2), V is permeate volume (m^3), and t is filtration time (s). Rejection mechanism in microfiltration is molecular sieving, thus flow resistances in microfiltration are membrane resistance (R_m) and cake layer resistance (R_c). Flux of solution is rearranged to Eq.2.5.

$$J_V = \frac{\Delta P}{((\mu_V \cdot (R_M + R_C))} \tag{2.5}$$

where J_v is flux of solution ($m^3/m^2 \cdot h$), A is membrane area (m^2), μ_v is viscosity of solution (Pa·s), ΔP is differential pressure between cake layer and membrane surface (Pa), R_m and R_c are membrane resistance and cake layer resistance (m^{-1}), respectively. The rejection percentage of MF membrane is defined in Eq.2.6.

$$R = \left(1 - \left(\frac{C_P}{C_R}\right)\right) \cdot 100 \tag{2.6}$$

where R is percentage of rejection, C_P and C_R are concentrations of solutes in permeate and retentate (mg/L), respectively.

Microfiltration can be operated in both dead-end and cross-flow modes. In dead-end mode, feed solution flows in tangential with membrane surface and retained particles accumulate and form a type of cake layer on the membrane surface. The accumulation of cake layer on surface causes increasing membrane resistance and flux of solution decrease rapidly. Thus dead-end filtration is normally used to separate small particles in solution and operated in batch process. For crossflow filtration, feed solution flows in parallel direction with membrane surface and it will remove the accumulated particles on surface by scouring. Flux of solution does not decrease rapidly when compared with dead-end filtration. Permeate fluxes for dead-end and crossflow filtration are shown in Fig. 2.12.

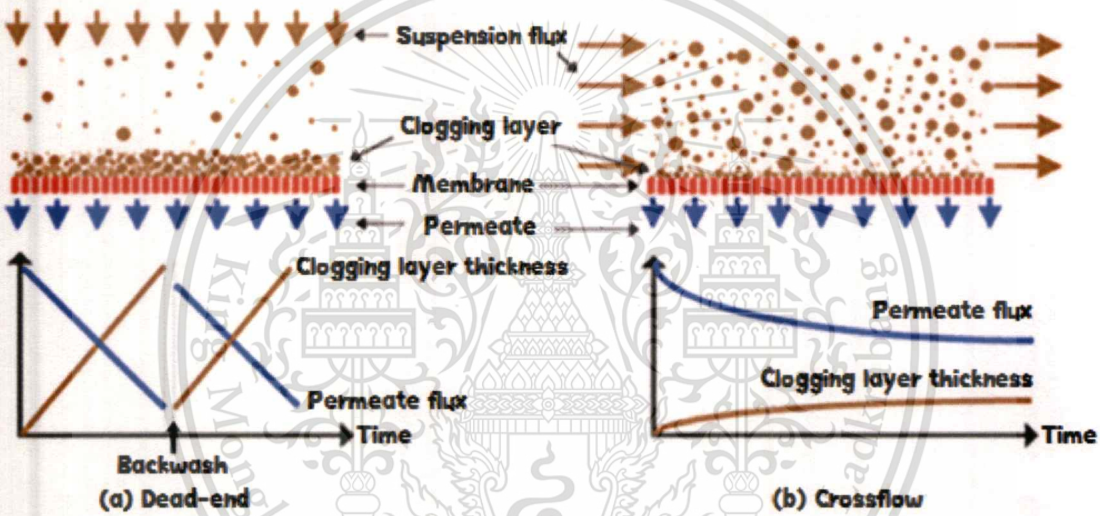


Figure 2.12 Schematic of solution flux in (a) dead-end and (b) crossflow filtration[19]

2.2.7 Ultrafiltration

Ultrafiltration (UF) is a low pressure (100-800 kPa or 1-8 atm) process for separating colloid, suspended particles and macromolecules with diameters in a range of 0.01-0.1 μm (or molecular weight cut-off of 20,000-100,000 daltons) by microporous membranes. Figure 2.13 shows the schematic of ultrafiltration characteristic.

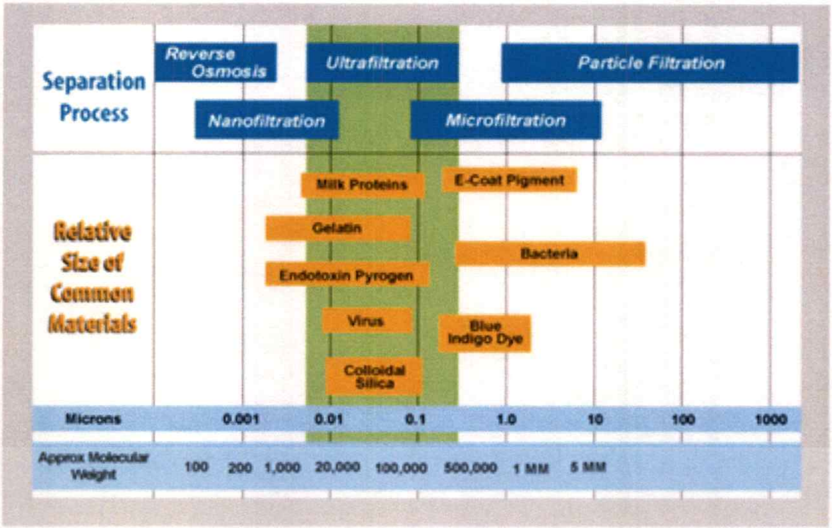


Figure 2.13 Schematic diagram of retained particles size and MWCO on ultrafiltration[20]

Ultrafiltration can be operated in both of dead-end and cross-flow filtration like in microfiltration. Rejection mechanism of UF membrane is molecular sieving by interaction between solutes and membrane, flow resistance is total flow resistance. Flux of solution for UF membrane is calculated by Eq.2.7.

$$J_v = \frac{(\Delta P - \Delta \pi)}{\mu_v \cdot R_t} \tag{2.7}$$

Where J_v is flux of solution ($m^3/m^2{\cdot}s$), ΔP and $\Delta\pi$ are differential of applied pressure and osmotic pressure (Pa), μ_v is viscosity of solution (Pa•s) and R_t is total flow resistance (m^{-1}), respectively. For solution with large size particles, value of $\Delta\pi$ is less than ΔP ($\Delta\pi << \Delta P$), thus exclude $\Delta\pi$ term and rearrange to Eq.2.8.

$$J_v = \frac{\Delta P}{\mu_v \cdot R_t} \tag{2.8}$$

The rejection percentage of UF membranes is defined in Eq.2.9.

$$R = \left(1 - \left(\frac{C_P}{C_R}\right)\right) \cdot 100 \tag{2.9}$$

where R is percentage of rejection, C_P and C_R are concentrations of solutes in permeate and retentate (mg/L), respectively.

2.3 Chitosan[6,21-25]

Chitosan is a copolymer composed of poly [β-(1,4)-2-amino-2-deoxy-D-glucopyranose] and poly [β-(1,4)-2-acetamido-2-deoxy-D-glucopyranose]. It is obtained from partially alkaline deacetylation of chitin and also found naturally in crustacean shell. Chitosan is regarded as chitin which acetyl group at C-2 position is replaced by amino group. Acetyl group in chitin structure is changed to amino group in deacetylation process and it refers to chitosan when degree of deacetylation (DD) reaches 50%. Figure 2.14 shows the structure of partially deacetylation of chitin and chitosan, whereas Fig. 2.15 shows the structure of chitin and chitosan.

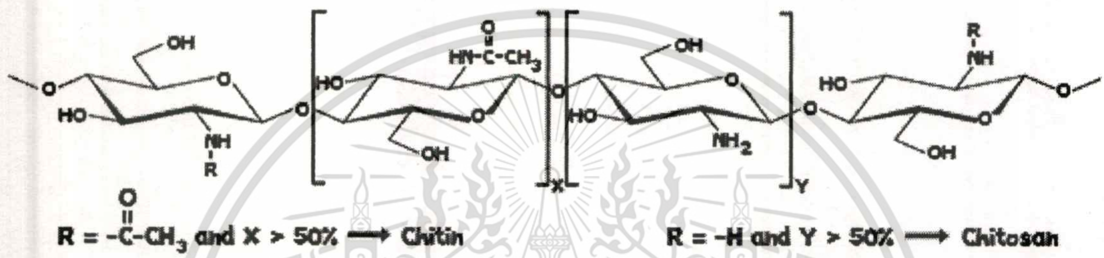


Figure 2.14 Structure of partially deacetylation of chitin and chitosan[6]

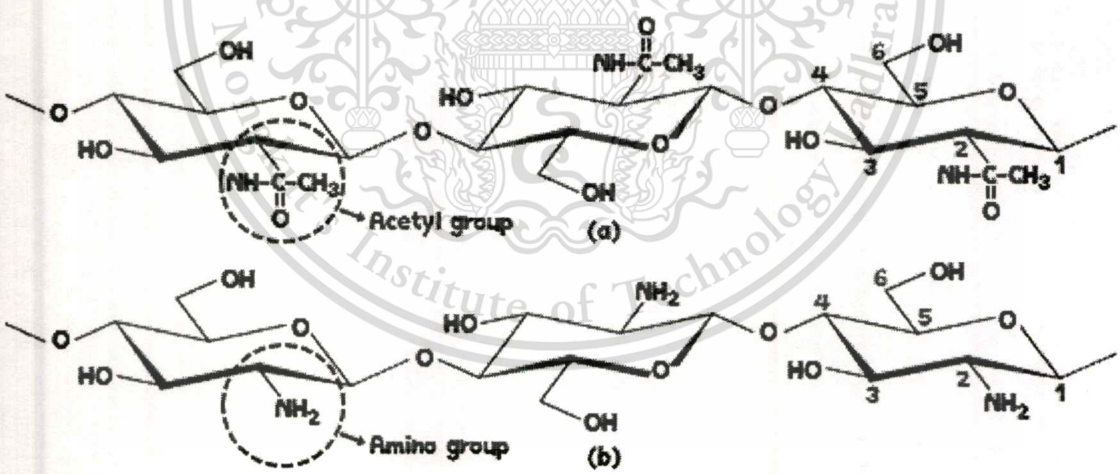


Figure 2.15 Structures of (a) chitin and (b) chitosan[21]

2.3.1 Production of chitosan[6]

Chitosan are obtained from the alkaline deacetylation of chitin. Chitin is extracted from crustacean shell wastes. Extraction procedure is divided into 3 steps including 1) deproteinization,

2) demineralization, and 3) decoloration. The productions of chitin and chitosan are shown in Fig. 2.16.

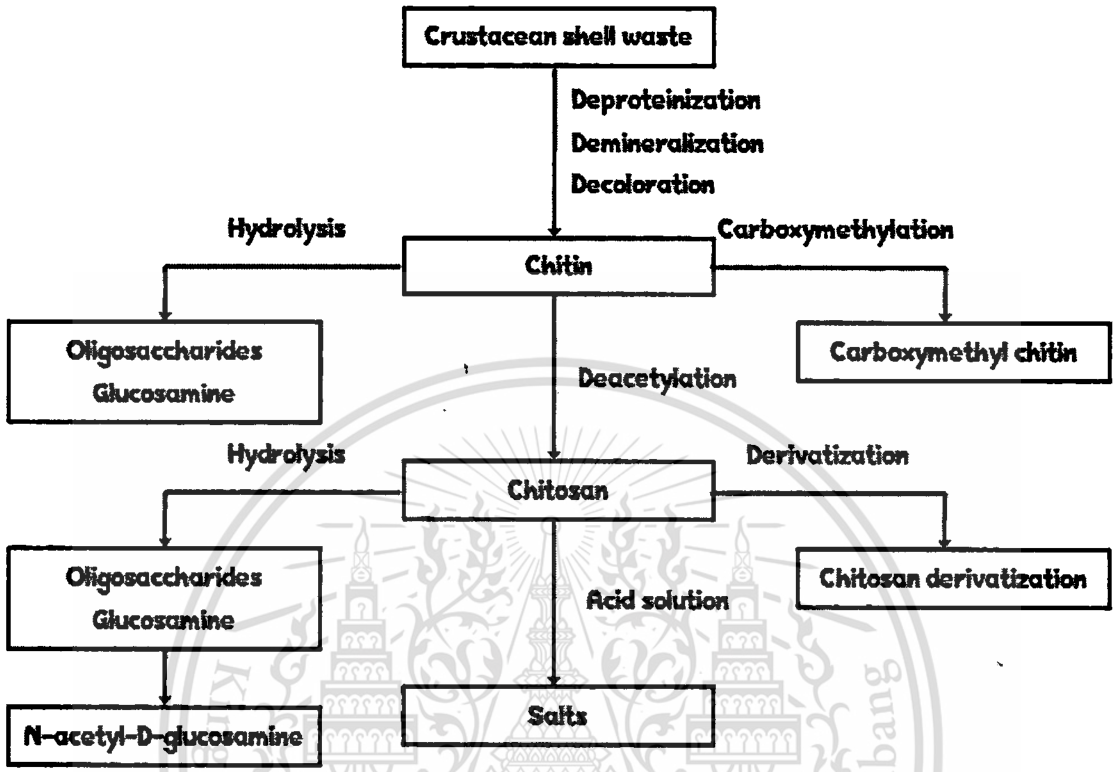


Figure 2.16 Simplified representation of preparation of chitin, chitosan and their derivatives[22]

In the production of chitin, crustacean shell wastes are extracted for proteins before demineralization and decoloration process because these proteins have a good quality than the products obtained after demineralization and decoloration process. First step, shellfish source is deproteinized by grinding, immersing and mixing thoroughly in 1-10% (wt.) NaOH at 65-100°C. Mixing time of shell wastes in alkaline solution depends upon the conditions in the extraction procedure. If this procedure takes a long-term period in strong conditions, chitin chains are depolymerized. After deproteinization process, crustacean shell wastes are demineralized by mixing thoroughly in dilute HCl solution (more than 0.7 M) at room temperature for 2-3 hours. The demineralization process removes minerals in shell wastes. In decoloration process, crustacean shell wastes are bleached by bleaching powder such as ethanol (C_2H_5OH), sodium hypochlorite (NaOCl), acetone (C_3H_6O), and 3% hydrogen peroxide (H_2O_2). After extraction process, chitin is washed and dried.

Chitosan is chemical derivative obtained by alkaline deacetylation of chitin. In deacetylation process, chitin is immersed in 40-50% (wt.) NaOH or KOH at high temperature (≥ 100°C) for 1-3 hours. After deacetylation process, chitosan is washed and dried. Figure 2.17 shows the deacetylation process of chitin.

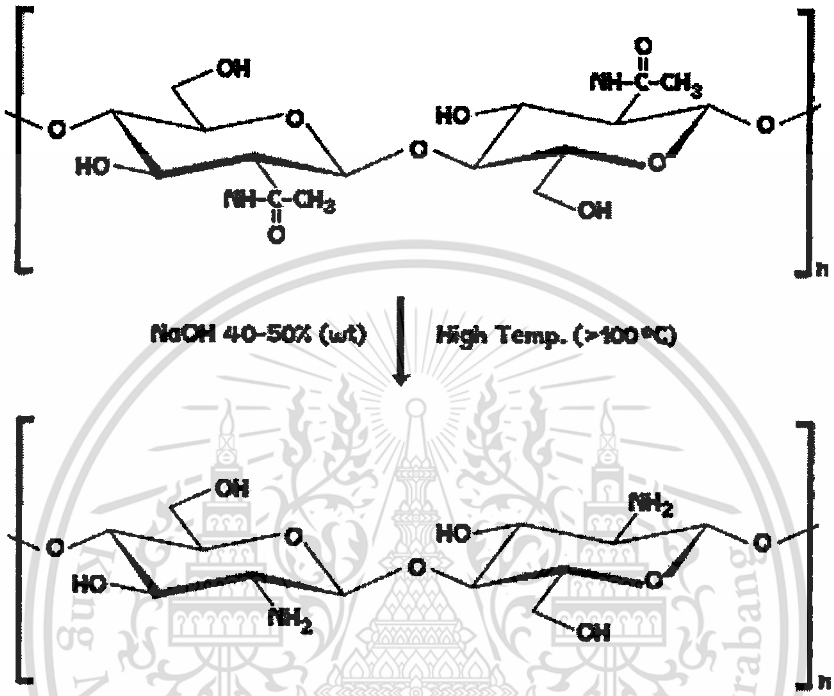


Figure 2.17 Deacetylation process of chitin[23]

2.3.2 Properties of chitosan[6,21-25]

2.3.2.1 Solubility of chitosan

Chitosan is normally insoluble in water at near neutral pH and most common organic solvent, but it is readily soluble in dilute acidic solutions (acetic acid, formic acid, hydrochloric acid, lactic acid, and inorganic acids with the remarkable exception of sulphuric acid) at pH below 6.0. As the pKa value of amino group on chitosan structure is 6.3, at low pH (≤ 6.0) these amino get protonated and become positively charged. That makes chitosan as a water-soluble cationic polyelectrolyte. On the other hand, as pH increases above 6.0, these amine groups become deprotonated, causing chitosan loses its charge and becomes insoluble. The dissolution constant (K_a) of the amine group is obtained from the equilibrium as shown in Eq.2.10.

$$-NH_2 + H_2O \rightleftharpoons -NH_3^+ + OH^- \quad (2.10)$$

2.3.2.2 Degree of acetylation (DA) and degree of deacetylation (DD)

Chitin and chitosan are poly [β-(1,4)-2-acetamido-2-deoxy-D-glucopyranose] and poly [β-(1,4)-2-amino-2-deoxy-D-glucopyranose], respectively. Degree of acetylation (DA) is a parameter defined as a mole fraction of acetylated units in the polymer chain. Chitin refers to high DA polymers (ideally 100% DA), whereas chitosan refers to low DA polymers (ideally 0% DA). When the DA reaches 50% or lower, chitin becomes chitosan due to the protonation of amine groups in glucosamine units. Chitin has a degree of acetylation (DA) of typically 0.90, indicating that the presence of some amine groups (as some amount of deacetylation might take place during extraction, chitin may also contain about 5-15% amine groups), while chitosan has a typical DA of less than 0.35.

Degree of deacetylation (DD) is a parameter defined as a mole fraction of deacetylated units in the polymer chain. Chitosan is referring to high DD polymers while chitin is referring to low DD polymers. Chitosan normally employs 50-95% DD. The correlation between DA and DD is shown in Eq.2.11.

$$\text{Degree of deacetylation (DD)} = 100 - \text{Degree of acetylation (DA)} \quad (2.11)$$

2.3.2.3 Molecular weight

Molecular weight of chitosan is calculated from intrinsic viscosity using the Mark-Houwink relation[21] as shown in Eq.2.12.

$$\eta = KM^{\alpha} \quad (2.12)$$

Where η is intrinsic viscosity, M is the molecular weight of chitosan, K and α are constant (depending on the nature of solvent and polymer). The values of K and α are listed in Table 2.4.

Table 2.4 The Mark-Houwink parameters for chitosan in various solvents[21]

Solvent	K (mg/L)	α	Temperature (°C)
0.1 M AcOH/0.2 M NaCl	1.81×10^{-3}	0.93	25
0.1 M AcOH/0.02 M NaCl	3.04×10^{-3}	1.26	25
0.2 M AcOH/0.1 M AcONa/4 M urea	8.93×10^{-2}	0.71	25
0.3 M AcOH/0.2 M AcONa (DA = 0.02)	8.2×10^{-2}	0.76	25
0.3 M AcOH/0.2 M AcONa (0 < DA < 0.03)	7.9×10^{-2}	0.796	25
0.02 M acetate buffer/0.1 M NaCl	8.43×10^{-2}	0.92	25

The weight average molecular weight of chitin and chitosan are 1.03×10^6 - 2.5×10^6 and 1×10^5 - 5×10^5, respectively.

2.3.2.4 Viscosity

Viscosity of chitosan solution depends on various factors, i.e. degree of acetylation, molecular weight, ionic strength, pH of chitosan solution and temperature. Generally, viscosity of chitosan decreases when increasing temperature. Type of acid solvent and pH of chitosan solution take effect on the viscosity. In acetic acid, viscosity of chitosan increases when decreasing pH but viscosity of chitosan in hydrochloric acid increases when increasing pH of chitosan solution.

2.3.2.5 Degradation

Chitin and chitosan are degradable and become short chain molecules as oligomer or oligosaccharide. Finally, these oligomers or oligosaccharides become monomers or monosaccharides of chitin and chitosan as N-acetyl-D-glucosamine and D-glucosamine.

2.3.3 Applications of chitosan

Chitosan has been widely used because it is harmless to human and presents excellent biological properties such as biodegradation, immunological, antibacterial and wound-healing activity[23]. This versatile material is used in food and beverages industry, agriculture, cosmetics and toiletries, biopharmaceutics, textile industry, paper production, and wastewater treatment process[6]. In addition, chitosan is also widely applied in water and wastewater treatment processes because it can be conditioned and used for pollutant complexation in different forms (gels, beads, solution, membranes, fibers etc.)[23]. Applications and properties of chitosan in water and wastewater treatment are shown in Table 2.5 and 2.6, respectively.

Table 2.5 Principle application of chitosan in industrial processes[21]

Food and beverages :	Not digestible by human (dietary fiber)
	Bind lipids (reduce cholesterol)
	Preservative
	Thickener and stabilizer for sauces
	Protective, fungistatic, antibacterial
Agriculture :	Defensive mechanism in plants
	Stimulation of plant growth
	Seed coating, frost protection
	Time release of fertilizers and nutrients into the soil
Cosmetics and toiletries :	Maintain skin moisture
	Treat acne
	Improve suppleness of hair
	Reduce static electricity in hair
	Oral care (toothpaste, mouthwashes, chewing gum)
Biopharmaceutics :	Immunologic and antitumoral
	Hemostatic and anticoagulant
	Healing and bacteriostatic
Textile industry :	Coating on fabric to reduce sweat and odors
Paper production :	Dry strength and wet strength agent in paper production
Wastewater treatment :	Coagulant and flocculant of interested molecules
	Removal of metal ions or dye
	Reduce odors

Table 2.6 Principle properties of chitosan in relation to its use in water and wastewater treatment application[24]

Principal characteristics	Potential application
Non-toxic	Flocculant to clarify water (drinking water)
Biodegradable	Reduction of turbidity in food processing effluents
Renewable resource	Coagulation of suspended solids, mineral and organic suspensions
Ecologically acceptable polymer (eliminating synthetic polymers, environmentally friendly)	Flocculation of bacterial suspensions
Efficient against bacteria, viruses and fungi	Interaction with negatively charged molecules
Formation of salts with organic and inorganic acids	Recovery of valuable products (i.e. protein)
Ability to form H-bonds intermolecularly	Chelation of metal ions
Ability to encapsulate	Removal of dye molecules by adsorption processes
Removal of pollutants (with pollutant-binding capacities)	Reduction of odours Sludge treatment Filtration and separation Polymer assisted ultrafiltration

2.4 Chitosan-enhanced membrane filtration[5]

Chitosan-enhanced membrane filtration is a process that combines between binding of interested molecules or particles to a water-soluble polymer and membrane filtration such as MF or UF membrane. MF and UF membranes are porous membranes, thus particles or molecules less than pore size can pass through the membrane. Chitosan is used for binding molecules or particles to form macromolecules larger than membrane pores size and those molecules can be retained by MF or UF membrane. Figure 2.18 shows the chitosan-enhanced membrane filtration process.

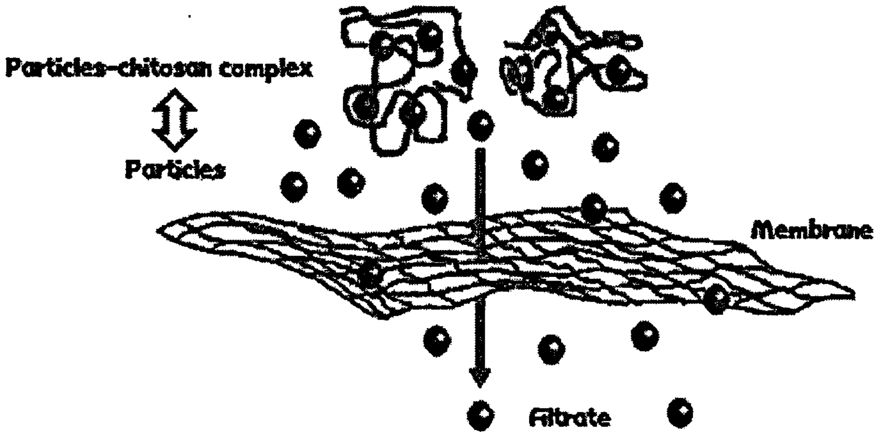


Figure 2.18 Chitosan-enhanced membrane filtration processes[25]

In this research, chitosan is used to enhance membrane filtration by crosslinking or formed covalent bond with formaldehyde in wastewater, and then chitosan combined with formaldehyde is retained by MF or UF membrane. Figure 2.19 shows the mechanism of chitosan and formaldehyde crosslinking.

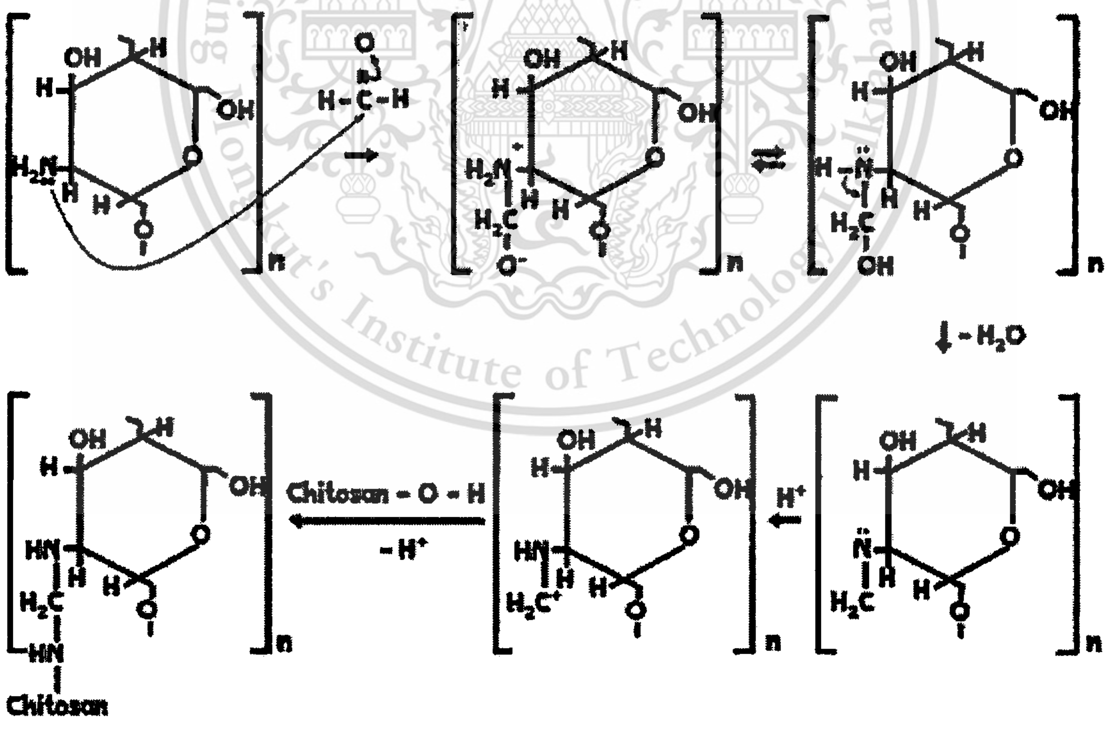


Figure 2.19 Mechanism of chitosan crosslinking by formaldehyde[26]

Figure 2.20 illustrated reaction between carbonyl group in formaldehyde molecule and amino group in chitosan structure.

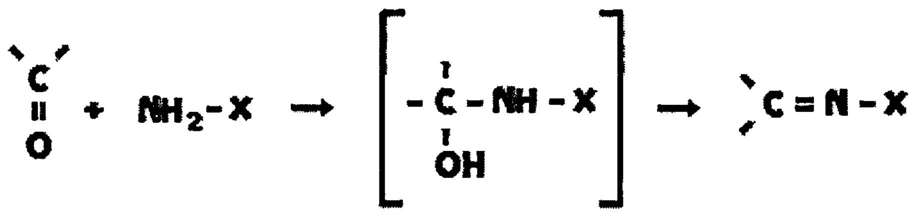


Figure 2.20 Reaction between carbonyl and amino group[27]

2.5 Literature reviews

Tanada *et al.* (1999)[2] studied on removal of formaldehyde by adsorption using activated carbon containing amino groups. Activated carbon was treated with concentrate nitric and sulfuric acids to introduce nitro groups on the surface, then nitro groups were reduced by iron powder and distilled in hydrochloric acid to introduce amino groups. Specific surface area and pore volume of activated carbon of U-AC, N1-AC and N2-AC were calculated by BET and Cranston-Inkley methods. Specific surface areas were 1119, 981 and 950 m^2/g and pore volume are 0.566, 0.436 and 0.424 ml/g, respectively. %N/C and % H/C are 0, 2.5 and 1.8% and 1.8, 2.3 and 2.3%, respectively. Amount of formaldehyde adsorbed on activated carbon surface increased with amination on the surface because of the increase of interaction between activated carbon surface and formaldehyde. The results showed that amount of formaldehyde adsorbed on activated carbon was N1-AC > N2-AC > U-AC.

Juang *et al.* (2000)[28] studied on removal of copper, cobalt, nickel, and zinc from aqueous solution by chitosan-enhanced membrane filtration. The experimental parameters such as pH of solutions, chitosan-metal ratio (β), applied pressure (ΔP), ionic strength on metal rejection were studied. Chitosan in this research was chitosan flake with 87% of degree of deacetylation and molecular weight 410 kDa. The regenerate cellulose ultrafiltration membrane, YM10 and YM30, with MWCO 10 and 30 kDa were used. The results showed that increasing of applied pressure increased solution flux (J_v) and decreased metal rejection (R_M), the constant J_v obtained at $\Delta P > 30$ psi and the metal rejection increased with increasing chitosan-metal ratio (β) but normalize flux (J_v/J_w) decreased with increasing β. At pH < 6, R_M slightly decreased and then increased by rising pH, which at pH > 8 showed complete R_M ($R_M = 1$). For example, the optimum conditions of Cu^{2+} removal ($R_M = 0.8$) were found to be a pH of 6 and $\beta = 3$ at $\Delta P = 30$ psi and metal concentration ($[M^{2+}]$) = 1.24 mM with J_v/J_w of 0.97 and 0.85 for YM10 and YM30 membranes, respectively. The effect of ionic strength was studied using inorganic salts (NaCl, $CaCl_2$ and Na_2SO_4). The results showed that J_v/J_w decreased by adding a small amount of NaCl and $CaCl_2$ but different effect in case of Na_2SO_4, i.e. increasing amount of Na_2SO_4 increased J_v/J_w at first and then slightly decreased.

Verbych *et al.* (2006)[29] studied on removal of copper from aqueous solution by means of ultrafiltration enhanced with chitosan. The experimental parameters, such as initial concentration of metal ions, background electrolytes, pH of solutions, ultrafiltration membranes of different molecular weight cut-offs, and polymer rejection, were studied. Chitosan used in this research was chitosan flake with molecular weight of 170 kDa. Cellulose-acetate membranes with MWCO 30 and 100 kDa were used. The initial Cu^{2+} ion concentration varied from 0.0078-0.469 mmol/L and chitosan concentration varied from 50-300 mg/L. The results showed that increasing pH of solution and chitosan:copper molar ratio (β) increased the rejection of Cu^{2+} ion but permeate flux decreased when increased pH of solution and chitosan:copper ratio. The most effective removal of copper ions (R_{Cu} = 98%) was achieved at pH 6 with β = 10 on a UAM-300 membrane. An increase of salt concentration resulted in the squeezing of a double-electric layer and deterioration of metal ion bonds with polymer functional groups. NaCl (1-1 type electrolyte) introduction into the solution did not practically influence Cu^{2+} ion retention, whereas in the presence of $CaCl_2$ (2-1 type electrolyte), removal of Cu^{2+} ion from the solution became worse.

Aroua *et al.* (2007)[30] studied on removal of chromium species from aqueous dilute solutions using polymer-enhanced ultrafiltration (PEUF) process. Polysulfone hollow fiber membrane with MWCO 500 kDa and three water-soluble polymer, chitosan, pectin, and PEI were used. The effects of pH and polymer composition on rejection coefficient and permeate flux at constant pressure have been investigated. The result show that pectin showed better performance for the separation of Cr (III) than chitosan and PEI. Whereas PEI showed better performance for the removal of Cr (VI) compared to the pectin and chitosan. Solution pH was found to be the major factor which controls the rejection of both chromium species. For Cr (III), high rejections approaching 100% were obtained at pH more than 7 for the three tested polymers. Percent retention of Cr (VI) remained almost constant around a value of less than 50% with chitosan and pectin, for PEI the retention of Cr (VI) approached 100% at low pH and sharply decreased at pH 9 and above. Permeate flux remained almost constant around 25% of pure water flux. However, the pectin systems showed slightly lower PEUF flux compared to chitosan and PEI. Effect of pectin and PEI concentrations on the retention of both chromium ions are also investigated. It was showed that percent rejection increased with change of pectin and PEI concentrations but reduced when the concentration is very high. For Cr (III), rejection was high and not much effected by the change of both pectin and PEI concentrations.

Barakat *et al.* (2010)[5] studied on removal of Cu(II), Ni(II), and Cr(III) from synthetic wastewater solutions by polymer-enhanced ultrafiltration process. Carboxy methyl cellulose was used for complexing the cationic forms of the heavy metals before filtration, then size of the complex has to be larger than the membrane pores and can be retained. A polyethersulfone ultrafiltration membrane with MWCO 10 kDa was used in the system and parameters affecting the percentage rejection of the metals, such as pH, metal ion concentration, CMC/metal ratio, and permeate flux have been investigated. Metal rejection in aqueous solution containing Ni(II), Cu (II) and Cr (III) ions in both of individuals and simultaneous solutions was also evaluated. The results showed that the complexation and filtration processes are pH dependent, the metal rejection was more efficient at neutral and alkaline conditions than at acidic. The results showed that the membrane had worked efficiently on a wide range of concentration up to 100 mg/L for both Cu (II) and Cr (III) ions, while rejection efficiency of Ni (II) ions decreased when concentration increased. Results obtained revealed that the maximum percentage of the metal rejection was achieved at pH $\geq$ 7 with increasing of the CMC concentration. The metal rejection efficiency values with initial metal ion concentration of 25 mg/L, CMC concentration of 1 g/L at pH 7 and pressure 1 bar were 98.6%, 99.1%, and 95.1% for Cu (II), Cr (III), and Ni (II), respectively for individuals solution. In simultaneous solutions, metal rejection efficiency were 98%, 98.3, and 94.4% for Cu (II), Cr (III), and Ni (II), respectively.

Haiqin Rong *et al.* (2010)[27] studied on a modified Rayon-based activated carbon fibers (ACFs) by p-aminobenzoic acid (PABA) and investigated their porous structure, surface chemistry, and formaldehyde removal behavior. The results showed that nitrogen-containing functional groups presented on the surface of modified Rayon ACFs and showed much higher adsorption capacity and longer breakthrough time for formaldehyde, formaldehyde removal by modified ACFs was attributed to the combined effects of physisorption contributed by pore structures and chemisorption contributed by the N-containing functional groups, whereas there was only physisorption between the as-prepared ACF and formaldehyde molecules.

CHAPTER 3

RESEARCH METHODOLOGY

3.1 Chemical

1. Acetylacetone ($C_5H_8O_2$) Analytical grade, Fluka, Switzerland.
2. Ammonium acetate (CH_3COONH_4) Analytical grade, Ajax Finechem Pty Ltd, Austria.
3. Chitosan flake, NNC Product Ltd, Thailand.
4. Citric acid monohydrate ($C_6H_8O_7 \cdot H_2O$) Analytical grade, Fluka, Switzerland.
5. Ethyl alcohol (CH_3CH_2OH) Analytical grade, Carlo Erba, Italy.
6. Formaldehyde solution 40% (CH_2O) Analytical grade, Carlo Erba, Italy.
7. Glacial acetic acid (CH_3COOH) Analytical grade, Lab Scan, Thailand.
8. Sodium hydroxide (NaOH) Analytical grade, Carlo Erba, Italy.
9. Sodium sulfite anhydrous (Na_2SO_3) Analytical grade, Carlo Erba, Italy.
10. Sodium metabisulfite ($Na_2S_2O_5$) Analytical grade, Carlo Erba, Italy.
11. Sulphuric acid 98% (H_2SO_4) Analytical grade, Mallinckrodt chemical, Germany.
12. Thymolphthalein indicator ($C_{28}H_{30}O_4$) Ajax Finechem Pty Ltd, Austria.

3.2 Apparatus

1. UV-Visible Spectrophotometer; T60 model, PG Instrument Limited, USA.
2. Total Organic Carbon Analyzer; TOC-V_{CSH}, Shimadzu, Japan.
3. pH meter; 827 pH Lab, Metrohm, Switzerland.
4. Turbidimeter; model 2100P, Hach. USA.
5. Orbital shaker, Gallenkamp, United Kingdom.
6. Temperature bath; WB 22, Memmert, England.
7. Filter paper 0.45 μm; Whatman, English.
8. Filter paper 5A; Toyo Roshi Kaisha Ltd, Japan.
9. Microfiltration and ultrafiltration unit
10. Microfiltration membrane; Polypropylene filter 5 μm, Kontec, USA.
11. Ultrafiltration membrane; UF-10B model, PURITY PRO ™ MEMBRANE, China.
12. Laboratory glassware

3.3 Experimental methods and analyses

All experiments were conducted in laboratory of Chemistry Department, Faculty of Science, King Mongkut's Institute of Technology Ladkrabang. The experiments were divided into 2 steps, including 1) batch tests of formaldehyde adsorption by chitosan and 2) chitosan-enhanced membrane filtration test, as shown in Fig. 3.1.

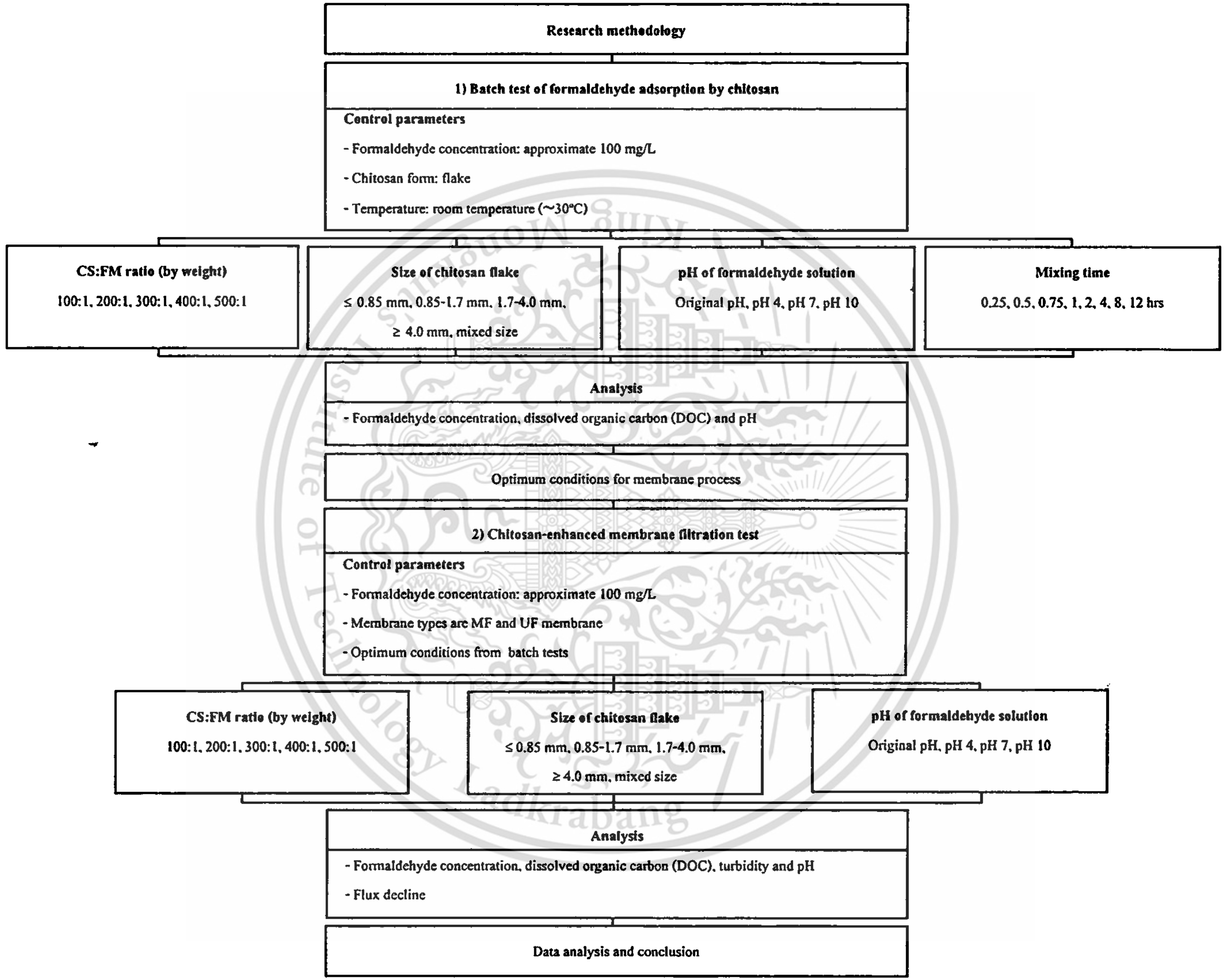


Figure 3.1 Steps of the research

3.3.1 Source of wastewater

Wastewater used in the experiment was formaldehyde synthetic wastewater. The synthetic wastewater was prepared from 40% (w/v) formaldehyde solution by diluting with RO water, initial concentration of formaldehyde was approximately 100 mg/L.

3.3.2 Batch tests of formaldehyde adsorption by chitosan

3.3.2.1) Chitosan form

Chitosan used in this research was mixed size chitosan flake with 85% deacetylation and molecular weight of 50,000 daltons.

3.3.2.2) Experimental set-up

Batch tests were conducted in 100-mL glass-amber bottles, wrapped outside with aluminum foil for protection of formaldehyde degradation as shown in Fig. 3.2.

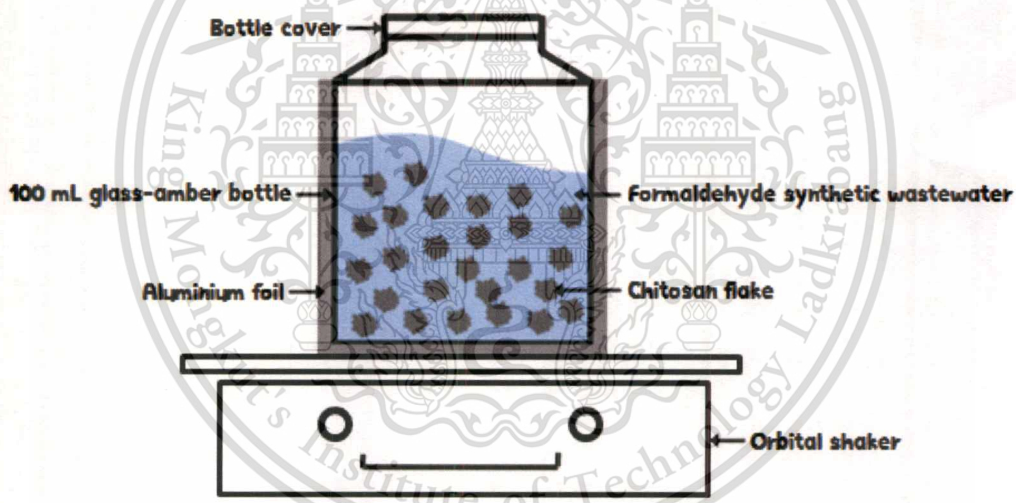


Figure 3.2 Schematic representation of batch test experimental set-up

3.3.2.3) Operating conditions and variable parameters

All batch experiments were performed at room temperature. The pH of initial formaldehyde was measured before and after mixing with chitosan. Factors affecting of formaldehyde adsorption by chitosan, i.e. chitosan:formaldehyde ratio (by weight), size of chitosan flake, pH of formaldehyde solution, and mixing time, were investigated. The factors affecting formaldehyde adsorption by chitosan batch tests are listed in Table 3.1.

Table 3.1 Variable parameters for batch test experiment

Variable parameters	Unit	Values
Preliminary test (mixing time)	hrs	12, 24
CS:FM ratio (by weight)	mg/mg	100:1, 200:1, 300:1, 400:1, 500:1
Size of chitosan flake	mm	≤ 0.85, 0.85-1.7, 1.7-4.0, ≥ 4.0 , mixed size
pH of formaldehyde solution	-	Original pH, pH 4, pH 7, pH 10
Mixing time	hrs	0.25, 0.5, 0.75, 1, 2, 4, 8, 12

1) Preliminary test

Preliminary tests on formaldehyde adsorption by chitosan were investigated to roughly determine adsorption time. 50 ml of formaldehyde synthetic wastewater with approximately concentration of 100 mg/L were mixed with chitosan flake 2.5 g in 100-mL glass-amber bottles, equivalent to 500:1 chitosan:formaldehyde ratio. Chitosan flake was used at mixed size and set of experiment without chitosan was conducted as a control group. Formaldehyde synthetic wastewater and chitosan were mixed using orbital shaker for 12 and 24 hours, then filtered through 0.45-μm filter papers prior to analyses of formaldehyde concentration, dissolved organic carbon (DOC), and pH.

2) Effect of chitosan:formaldehyde ratio

The effect of chitosan:formaldehyde ratio on formaldehyde adsorption by chitosan was investigated. 50 ml of formaldehyde synthetic wastewater with approximately concentration of 100 mg/L were mixed with chitosan flake of 0.5, 1.0, 1.5, 2.0, and 2.5 g in 100-mL glass-amber bottles, equivalent to 100:1, 200:1, 300:1, 400:1, and 500:1 chitosan:formaldehyde weight ratio. In this experiment chitosan flake of mixed size was used and a set of experiment without chitosan was conducted as a control group. Formaldehyde synthetic wastewater and chitosan were mixed using orbital shaker (mixing time was obtained from experiment 3.1), then filtered through 0.45-μm filter papers prior to analyses of formaldehyde concentration, dissolved organic carbon (DOC), and pH.

3) Effect of chitosan size

The effect of chitosan size on formaldehyde adsorption by chitosan was investigated. 50 ml of formaldehyde synthetic wastewater with approximately concentration of 100 mg/L were mixed with chitosan flake in 100-mL glass-amber bottles (chitosan:formaldehyde ratio was obtained from experiment 3.2), chitosan flake was used at size ≤ 0.85 mm, 0.85-1.7 mm, 1.7-4.0 mm, ≥ 4.0 mm, and mixed size. A set of experiment without chitosan was conducted as a control group. Formaldehyde synthetic wastewater and chitosan were mixed using orbital shaker (mixing time was obtained from experiment 3.1), then filtered through 0.45-μm filter papers prior to analyses of formaldehyde concentration, dissolved organic carbon (DOC), and pH.

4) Effect of pH of formaldehyde solution

The effect of pH of formaldehyde solution on formaldehyde adsorption by chitosan was investigated. 50 ml of formaldehyde synthetic wastewater with approximately concentration of 100 mg/L were mixed with chitosan flake in 100-mL glass-amber bottles (chitosan:formaldehyde ratio and size of chitosan flake were obtained from experiment 3.2 and 3.3). Formaldehyde synthetic wastewater was varied at original pH, pH of 4, 7, and 10 by adjusting with 1 M HCl or 1 M NaOH. A set of experiment without chitosan was conducted as a control group. Formaldehyde synthetic wastewater and chitosan were mixed using orbital shaker (mixing time was obtained from experiment 3.1), then filtered through 0.45-μm filter papers prior to analyses of formaldehyde concentration, dissolved organic carbon (DOC), and pH.

5) Effect of mixing time

The effect of mixing time on formaldehyde adsorption by chitosan was investigated. 50 ml of formaldehyde synthetic wastewater with approximately concentration of 100 mg/L were mixed with chitosan flake in 100-mL glass-amber bottles, (chitosan:formaldehyde ratio, size of chitosan flake and pH of formaldehyde solution were obtained from experiment 3.2, 3.3 and 3.4). A set of experiment without chitosan was conducted as a control group. Formaldehyde synthetic wastewater and chitosan were mixed using orbital shaker for 0.25, 0.5, 0.75, 1, 2, 4, 8 and 12 hours, then filtered through 0.45-μm filter papers prior to analyses of formaldehyde concentration, dissolved organic carbon (DOC), and pH.

3.3.3 Membrane processes for formaldehyde removal by chitosan-enhanced membrane filtration

3.3.3.1) Experimental set-up

Membrane processes in this study were divided into 2 parts including MF and UF units, Fig. 3.3 and 3.4 show schematic diagrams of MF and UF units, respectively.

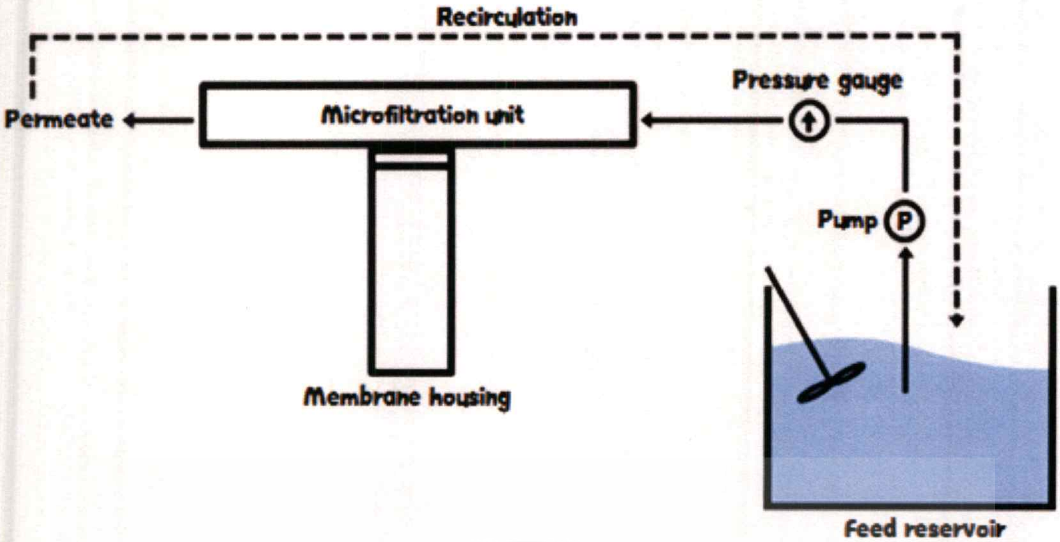


Figure 3.3 Schematic diagram of MF unit

MF unit was composing of polypropylene (PP) cartridge with pores size of 5 μm and filtration area of 0.013 m^2 located inside membrane housing. The MF unit was operated in dead-end mode. When MF unit was operated, feed solution containing formaldehyde was mixed thoroughly with chitosan by an agitator and delivered to membrane housing. An aliquot of permeate was collected for analysis and the remaining was recirculated to feed reservoir to make a constant volume of feed.

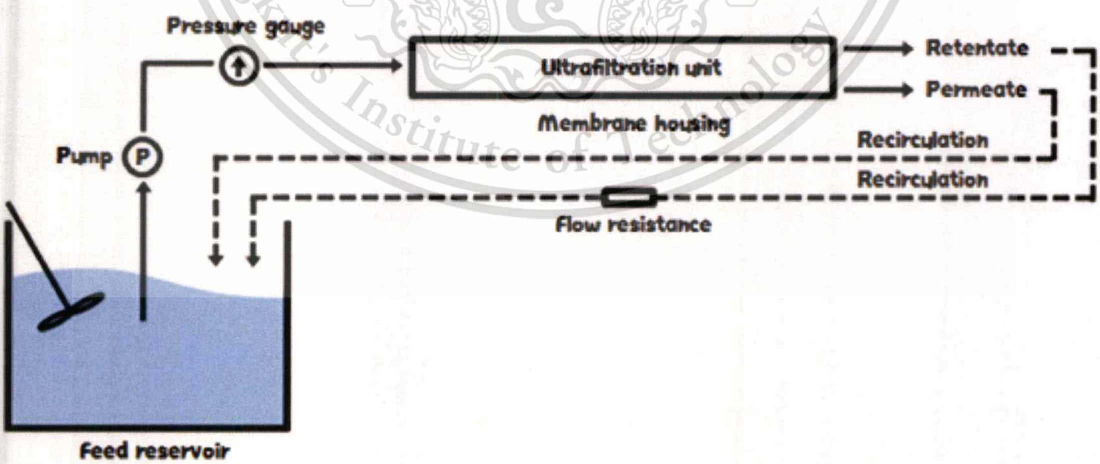


Figure 3.4 Schematic diagram of UF unit

UF unit was composing of polypropylene (PP) hollow-fiber with pore size of 0.01 μm and filtration area of 0.85 m^2 located inside membrane housing. The UF unit was operated in crossflow mode, creating permeate and retentate streams. When the UF unit was operated, feed solution containing formaldehyde was mixed thoroughly with chitosan flake by an agitator and fed to membrane housing. Aliquot of permeate and retentate were collected for analysis, then both streams were reservoir to make feed volume constant. Equipments used in this process are summarized in Table 3.2.

Table 3.2 Equipments for membrane processes

Equipment	Detail
Feed/Permeate reservoir	80 L polyethylene
Influent pump	Flow rate 0.6 L/min
Pressure gauge	80 psi
Agitator	Aluminum propeller
Permeate/retentate line	Teflon tube
MF membrane	Polypropylene cartridge
UF membrane	Polypropylene hollow-fiber

Before membrane operation, MF and UF membranes were cleaned by immersion in water for 15 minutes and placed in 0.0001 M citric acid ($C_6H_8O_7.H_2O$) solution for 15 minutes then rinsed with water for 15 minutes. After that, the membranes were placed in 0.001 M sodium hydroxide (NaOH) solution for 15 minutes and then rinsed with water for 15 minutes.

3.3.3.2) Operating conditions and variable parameters

The factors affecting membrane performance are investigated as listed in Table 3.3. Optimum condition, i.e. mixing time, was obtained from batch test. Before operation, membranes were flushed with water until water flux (J_w) became constant by determining volume of permeate per membrane area per time in a unit of $m^3/m^2 \cdot hr$. Permeate flux was determined in the same unit in order to investigate membrane fouling.

Table 3.3 Operating conditions and variable parameters for MF and UF membrane processes

Control parameters	Variable parameter
Membrane type : MF membrane **Chitosan form :** flake **Mixing time :** from batch test **Temperature :** room temperature (~30°C)	**CS:FM ratio (by weight):** 100:1, 200:1, 300:1, 400:1, 500:1 **Size of chitosan flake:** ≤ 0.85, 0.85-1.7, 1.7-4.0, ≥ 4.0 , mixed size **pH of formaldehyde solution:** original pH, pH 4, pH 7, pH 10
Membrane type : UF membrane **Chitosan form :** flake **Mixing time :** from batch test **Temperature :** room temperature (~30°C)	**CS:FM ratio (by weight):** 100:1, 200:1, 300:1, 400:1, 500:1 **Size of chitosan flake:** ≤ 0.85, 0.85-1.7, 1.7-4.0, ≥ 4.0 , mixed size **pH of formaldehyde solution:** original pH, pH 4, pH 7, pH 10

1) Effects of chitosan:formaldehyde ratio

The effects of chitosan:formaldehyde ratio on formaldehyde removal and flux decline by chitosan-enhanced membrane filtration were investigated. The optimum condition, i.e. mixing time, was obtained from batch tests. 10 liters of formaldehyde synthetic wastewater with approximately concentration of 100 mg/L were mixed with chitosan flake 100, 200, 300, 400, and 500 g in feed tank, equivalent to 100:1, 200:1, 300:1, 400:1, and 500:1 chitosan:formaldehyde ratio and delivered to membrane housing. Permeate and retentate was collected every 30 minutes and permeate flux was measured every 15 minutes. Feed, permeate, and retentate were analysed for formaldehyde concentration, dissolved organic carbon (DOC), turbidity, and pH.

2) Effects of chitosan size

The effects of chitosan size on formaldehyde removal and flux decline by chitosan-enhanced membrane filtration were investigated. The optimum condition, i.e. mixing time, was obtained from batch tests. 10 liters of formaldehyde synthetic wastewater with approximately concentration of 100 mg/L were mixed with chitosan flake in feed tank and delivered to membrane housing (chitosan:formaldehyde ratio was obtained from experiment 2.1), chitosan flake was used at size ≤ 0.85 mm, 0.85-1.7 mm, 1.7-4.0 mm, ≥ 4.0 mm, and mixed size.

Permeate and retentate were collected every 30 minutes and permeate flux was measured every 15 minutes. Feed, permeate, and retentate were analysed for formaldehyde concentration, dissolved organic carbon (DOC), turbidity, and pH.

3) Effects of pH of formaldehyde solution

The effects of pH of formaldehyde solution on formaldehyde removal and flux decline by chitosan-enhanced membrane filtration were investigated. The optimum condition, i.e. mixing time, was obtained from batch tests. 10 liters of formaldehyde synthetic wastewater with approximately concentration of 100 mg/L were mixed with chitosan flake in feed tank and delivered to membrane housing (chitosan:formaldehyde ratio and size of chitosan flake were obtained from experiments 2.1 and 2.2), formaldehyde synthetic wastewater was varied at original pH, pH of 4, 7, and pH 10 by adjusting with 1 M HCl or 1 M NaOH. Permeate and retentate were collected every 30 minutes and permeate flux was measured every 15 minutes. Feed, permeate, and retentate were analysed for formaldehyde concentration, dissolved organic carbon (DOC), turbidity, and pH.

3) Membrane cleaning

When membrane fouling occurred, cleaning of membrane was processed following these proceduces:

1) Hydrodynamic cleaning: fouled membrane was flushed with tap water in reverse direction of feed for 15 minutes then flushed with tap water in same direction of feed for 15 minutes.

2) Chemical cleaning: fouled membrane was flushed with tap water for 15 minutes and placed in 0.0001 M citric acid ($C_6H_8O_7.H_2O$) solution for 30 minutes then rinsed with water for 15 minutes. After that, fouled membrane was placed in 0.0001 M sodium hydroxide (NaOH) solution for 30 minutes and then rinsed with water for 15 minutes.

Before placing back to the membrane housing, cleaned membrane was flushed with water and water flux was measured. After membrane operation, MF and UF membranes were cleaned with the same procedure and placed in 1% (w/v) sodium metabisulfite ($Na_2S_2O_5$) to prevent the oxidation on membrane surface and kept in refrigerator at 4°C to prevent the growth of microorganisms on membrane surface.

3.3.4 Analyses and measurements

Parameters and methods of analyses that used in this study are given in Table 3.4.

Table 3.4 Measured parameters and methods

Parameters	Methods/ Equipments	References
Formaldehyde concentration	Colorimetric method, UV-Vis spectrophotometer	Nash, 1953[31]
Dissolved organic carbon (DOC)	Total organic carbon analyzer	APHA, 2005[32]
Turbidity	Turbidimeter	APHA, 2005[32]
pH	pH meter	APHA, 2005[32]

CHAPTER 4

RESULTS AND DISCUSSION

This research studied factors affecting treatment of synthetic wastewater containing formaldehyde using chitosan-enhanced membrane filtration. Batch tests were conducted to study the effects of chitosan:formaldehyde (CS:FM) ratio, chitosan size, pH of formaldehyde solution, and mixing time on chitosan adsorption performance. Optimums conditions obtained from batch were used as operating parameters in the operation of chitosan-enhanced membrane filtration. The performances of chitosan-enhanced MF and UF membranes filtration in removal of formaldehyde in synthetic wastewater were investigated.

4.1 Batch tests of formaldehyde adsorption by chitosan

4.1.1 Preliminary test

Preliminary test was conducted to roughly determine adsorption time for formaldehyde removal in batch test. The results showed that formaldehyde concentration in the solution decreased from 125.88 mg/L to 35.77 and 34.42 mg/L at 12 and 24 hours. Ability of formaldehyde adsorption by chitosan was constant which overall efficiencies in formaldehyde removal were in a range of 72-73%. Thus, in the next step of experiment, mixing time of 12 hours was chosen. When monitoring of dissolved organic carbon (DOC) in the solution, it was found that DOC in solution increased at 12 and 24 hours. It can be explained that the DOC remaining in the solution originated from the dissolution of organic carbon in chitosan molecules, leading to an increase of DOC and turbidity in the effluent.

According to the results from control groups and chemical properties of formaldehyde (i.e. high solubility, high boiling point, and low Henry's law constant), it was proven that most formaldehyde was absorbed by chitosan and only few amount of formaldehyde lost during experiment, possibly due to photodegradation or evaporation.

4.1.2 Effect of chitosan:formaldehyde ratio on formaldehyde removal

Chitosan:formaldehyde ratio at 100:1, 200:1, 300:1, 400:1, and 500:1 (by weight) were used in the experiment in order to investigate the effect of chitosan:formaldehyde ratio on adsorption of formaldehyde. The results showed that formaldehyde concentration in the solution decreased with increasing chitosan:formaldehyde ratio until 72% formaldehyde removal at chitosan:formaldehyde ratio of 500:1 (by weight), as illustrated in Fig. 4.1. The finding indicated an increase of chitosan in the solution provided more active sites to react with formaldehyde causing higher reduction of formaldehyde.

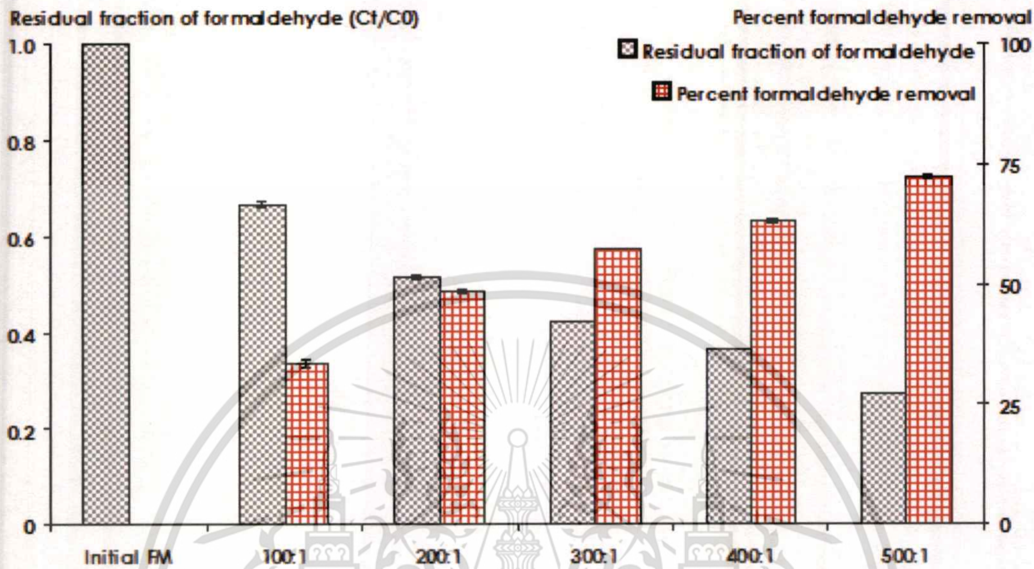


Figure 4.1 Residual fraction of formaldehyde and percent formaldehyde removal at different chitosan:formaldehyde ratio

(Chitosan flake of mixed size, original pH of solution, and mixing time of 12 hours)

However, it was found that DOC increased as chitosan:formaldehyde ratio increased because of more dissolution of chitosan that provided a large amount of DOC in the solution as illustrated in Fig. 4.2. According to the study of the effect of chitosan:formaldehyde ratio on formaldehyde removal, the highest chitosan:formaldehyde ratio used in this work was 500:1 (by weight). Because it provided the highest formaldehyde adsorption ability compared to other chitosan:formaldehyde ratio. Besides, amount of chitosan that increased with increasing chitosan:formaldehyde ratio generated a large amount of DOC in the solution and this consequence affected the quality of treated water. In the next step of experiment, the chitosan:formaldehyde ratio of 500:1 was chosen.

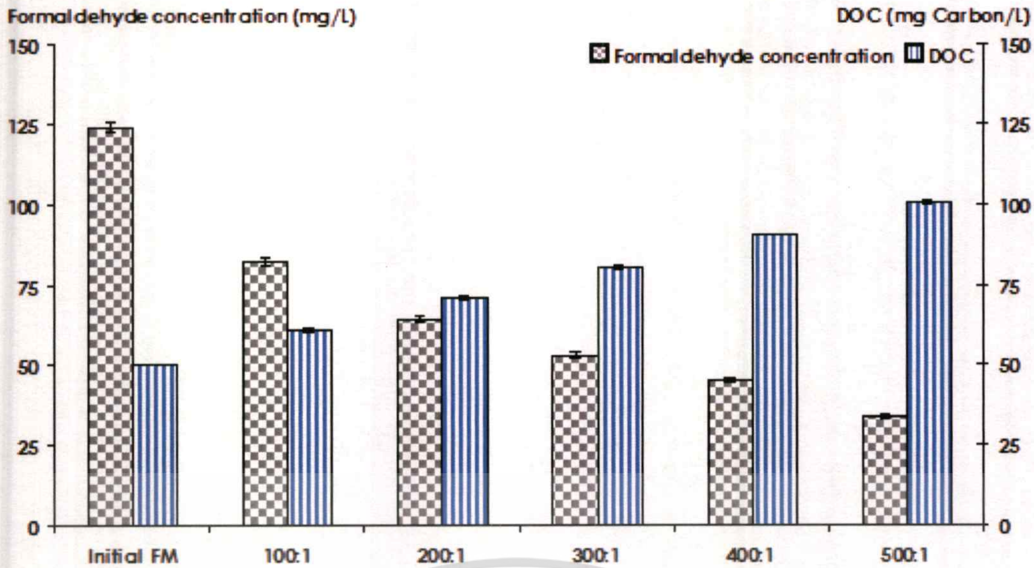


Figure 4.2 DOC levels of remaining in solution after chitosan adsorption at different chitosan:formaldehyde ratio

(Chitosan flake of mixed size, original pH of solution, and mixing time of 12 hours)

4.1.3 Effect of chitosan size on formaldehyde removal

Chitosan flake at different sizes were used in the experiment in order to investigate the effect of chitosan size on adsorption of formaldehyde. The results showed that formaldehyde removal using various sizes of chitosan flake were similar, as seen in Fig. 4.3. Almost formaldehyde adsorption by chitosan was chemisorptions by amino groups in chitosan molecules[2,27]. A reduction size of chitosan flake did not decrease amount of amino groups in chitosan molecules. Overall efficiency in formaldehyde removal was in a range of 71-72%.

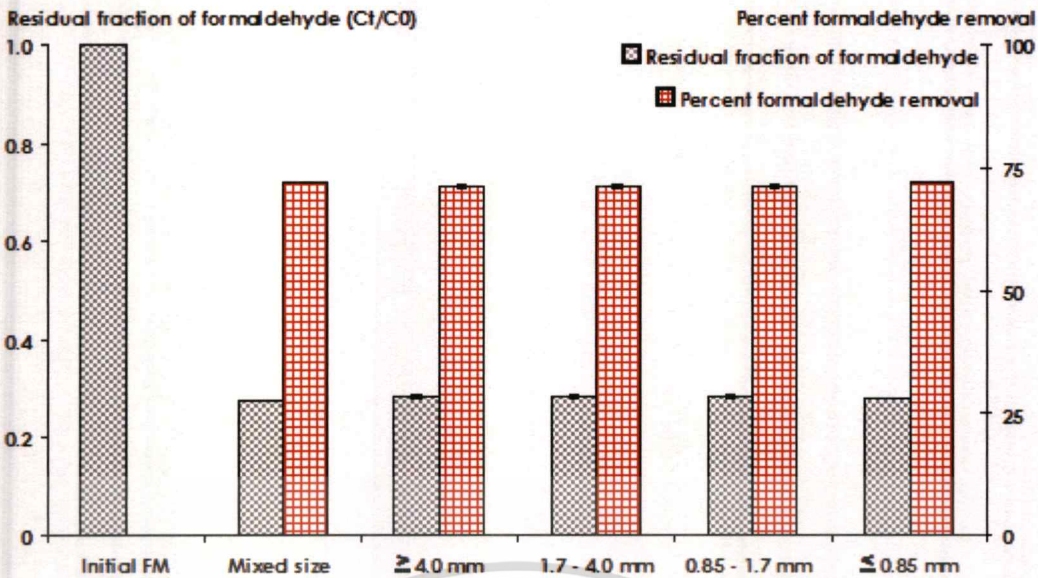


Figure 4.3 Residual fraction of formaldehyde and percent formaldehyde removal at different size of chitosan flake (chitosan:formaldehyde ratio of 500:1, and original pH of solution)

Figure 4.4 shows DOC of initial formaldehyde and remaining in solution, it was found that DOC remaining in the solution after adsorption were similar and slightly increased at chitosan sizes of 0.85-1.7 and ≤ 0.85 mm. It can be explained that the solubility of chitosan was slightly increased with chitosan size decreased, thus chitosan was more soluble and DOC in the solution after adsorption was increased. In the next step of experiment, mixed size chitosan was chosen.

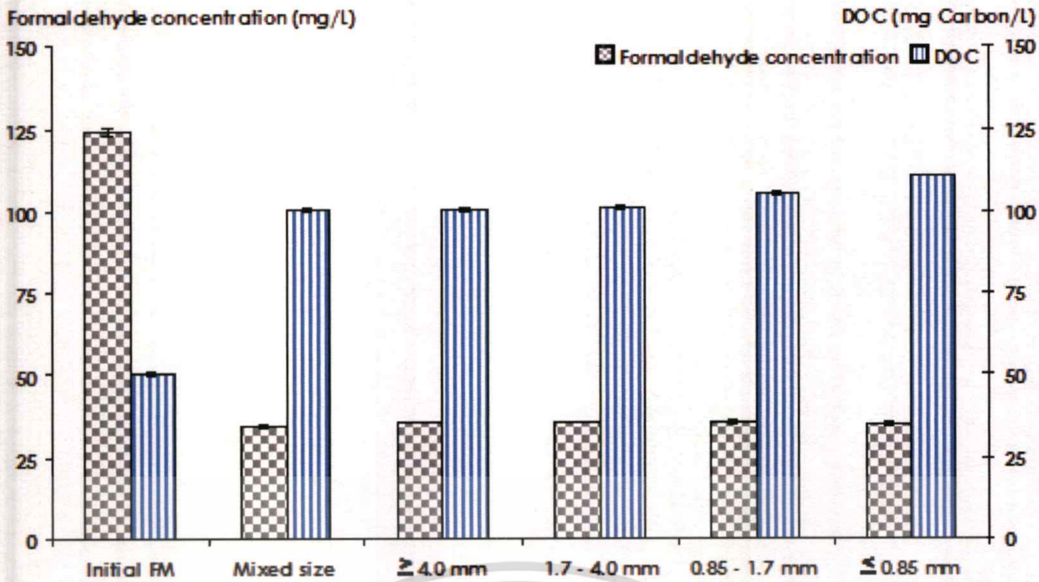


Figure 4.4 DOC levels of remaining in solution after chitosan adsorption

at different size of chitosan flake

(chitosan:formaldehyde ratio of 500:1, and original pH of solution)

4.1.4 Effect of pH of formaldehyde solution on formaldehyde removal

pH of formaldehyde solution at original pH and pH of 4, 7 and 10 were used in the experiment in order to investigate the effect of pH of formaldehyde solution on adsorption of formaldehyde. The results showed that there was no difference in formaldehyde removal using various pH of formaldehyde solution. As seen in Fig. 4.5, formaldehyde concentrations in the solution using different pH of formaldehyde solution were similar. Overall efficiency in formaldehyde removal was in a range of 71-72%. It can be explained that the reaction between chitosan and formaldehyde was crosslinking by formaldehyde as a crosslinking agent. When chitosan was mixed with formaldehyde solution, a part of chitosan that partial dissolved in the solution was crosslinked by formaldehyde at amino group of chitosan and oxygen atom of formaldehyde[26], while chitosan that did not dissolve in the solution and formed covalent bond with formaldehyde[27]. A variation in pH of solution did not affect crosslinking or covalent bond between chitosan and formaldehyde. Thus, percent removal of formaldehyde at different pH of solution was similar. In the next step of experiment, original pH of solution was chosen.

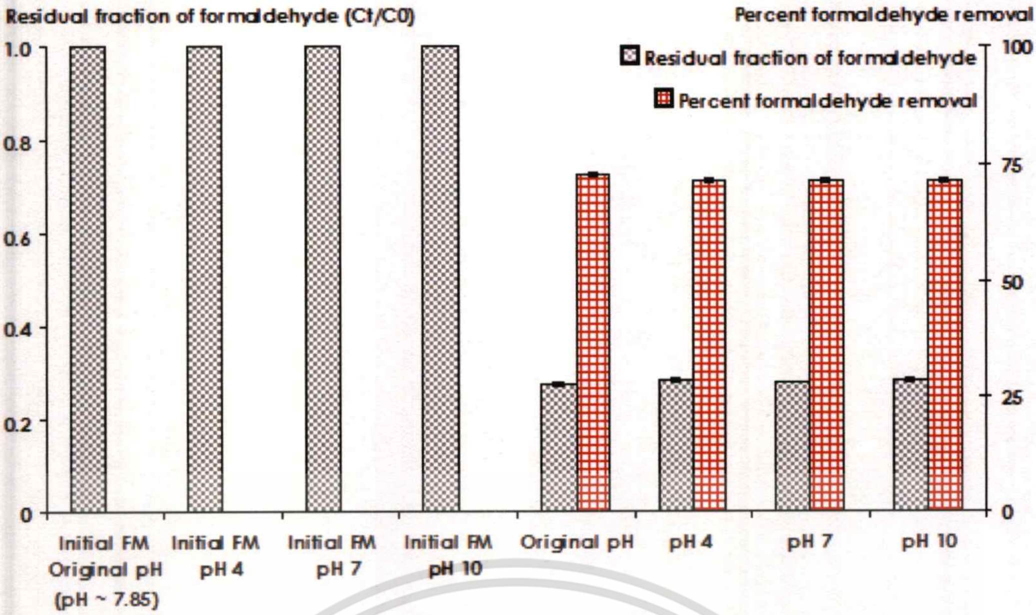


Figure 4.5 Residual fraction of formaldehyde and percent formaldehyde removal at different pH of formaldehyde solution

(chitosan:formaldehyde ratio of 500:1, chitosan flake of mixed size, and mixing time 12 hours)

Figure 4.6 shows DOC of initial formaldehyde and remaining in solution, it was found that DOC remaining in the solution after adsorption was similar at various pH of formaldehyde solution. It can be explained that pH of formaldehyde solution did not affect for the dissolution of chitosan, thus DOC remaining in solution after chitosan adsorption at different pH of formaldehyde solution were similar.

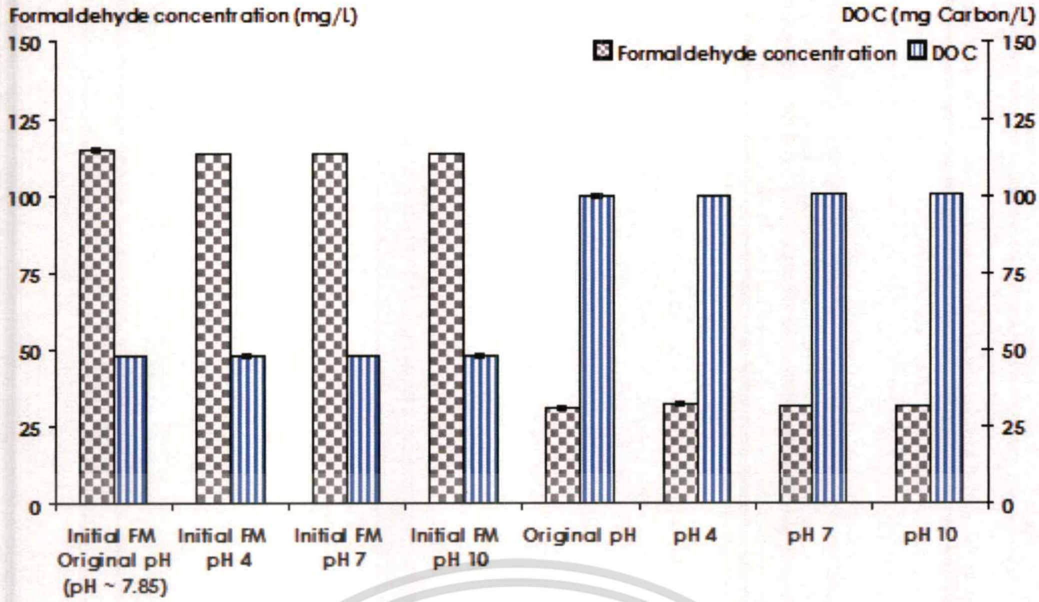


Figure 4.6 DOC levels of remaining in solution after chitosan adsorption at different pH of formaldehyde solution (chitosan:formaldehyde ratio of 500:1, chitosan flake of mixed size, and mixing time 12 hours)

When considering pH of solution after adsorption, it was found that pH of solution after adsorption was similar to initial pH. It can be explained that chemical bonding between chitosan and formaldehyde was covalent bond, proton and hydroxide in the solution did not affect for covalent bonding between chitosan and formaldehyde caused pH of the solution after adsorption was similar to initial pH[27], as illustrated in Fig. 4.7. In the next step of experiment, original pH of formaldehyde solution was chosen.

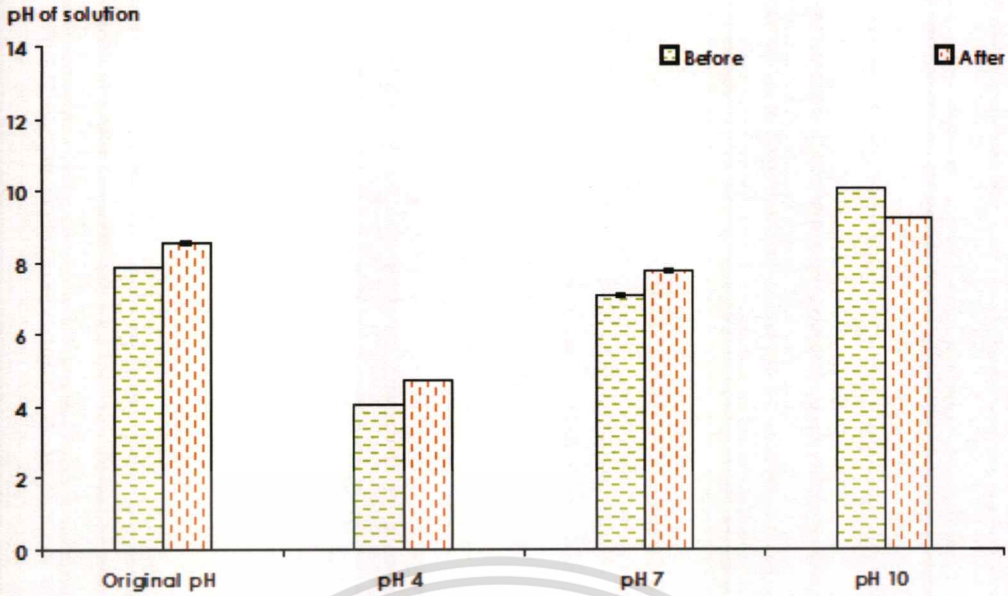


Figure 4.7 Comparison between pH of formaldehyde solution before and after mixed with chitosan

(chitosan:formaldehyde ratio of 500:1, chitosan flake of mixed size, and mixing time 12 hours)

4.1.5 Effect of mixing time on formaldehyde removal

Mixing time at 0.25, 0.5, 0.75, 1, 2, 4, 8 and 12 hours were used in the experiment in order to investigate the effect of mixing time on adsorption of formaldehyde. As seen in Fig. 4.8, formaldehyde concentration in the solution slightly decreased after 0.5 hours and almost stable after 4 hours. The results indicated that adsorption of formaldehyde by chitosan became saturated after 0.5 hours. Overall efficiency in formaldehyde removal was in a range of 71-72%. In the next step of experiment, mixing time 0.5 hours was selected.

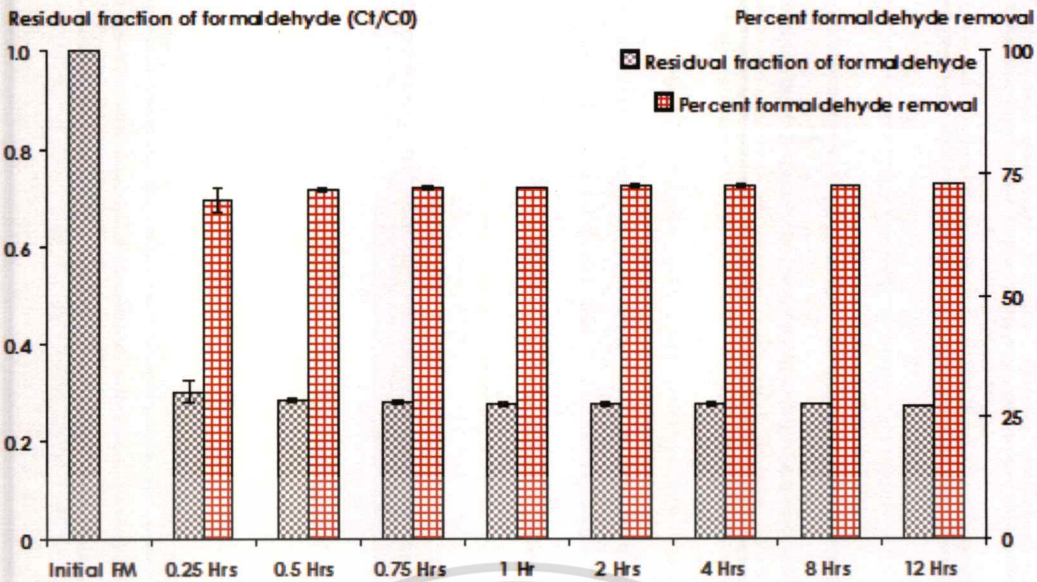


Figure 4.8 Residual fraction of formaldehyde and percent formaldehyde removal at different mixing time

(chitosan:formaldehyde ratio of 500:1, chitosan flake of mixed size, and original pH of solution)

4.2 Membrane processes for formaldehyde removal by chitosan-enhanced microfiltration (MF) and ultrafiltration (UF) membranes

4.2.1 Formaldehyde removal and flux decline by microfiltration (MF) and ultrafiltration (UF) membranes

In order to investigate efficiency of MF and UF membranes on formaldehyde removal and permeate flux, formaldehyde synthetic wastewater with approximately concentration of 100 mg/L without chitosan was used to operate the system. The results showed that formaldehyde removal efficiency of MF and UF membranes were similar and lower than 1%. It can be explained that molecular size of formaldehyde were smaller than pore size of MF and UF membranes, formaldehyde easily passed through pores of both membranes. On the other hand, formaldehyde was not retained by MF and UF membranes. Figure 4.9 shows normalized permeate flux (permeate flux at various time per initial flux) of MF and UF membranes, which was close to 1. It indicated that there was no fouling due to no solute clogged on membrane surface or in membrane pores.

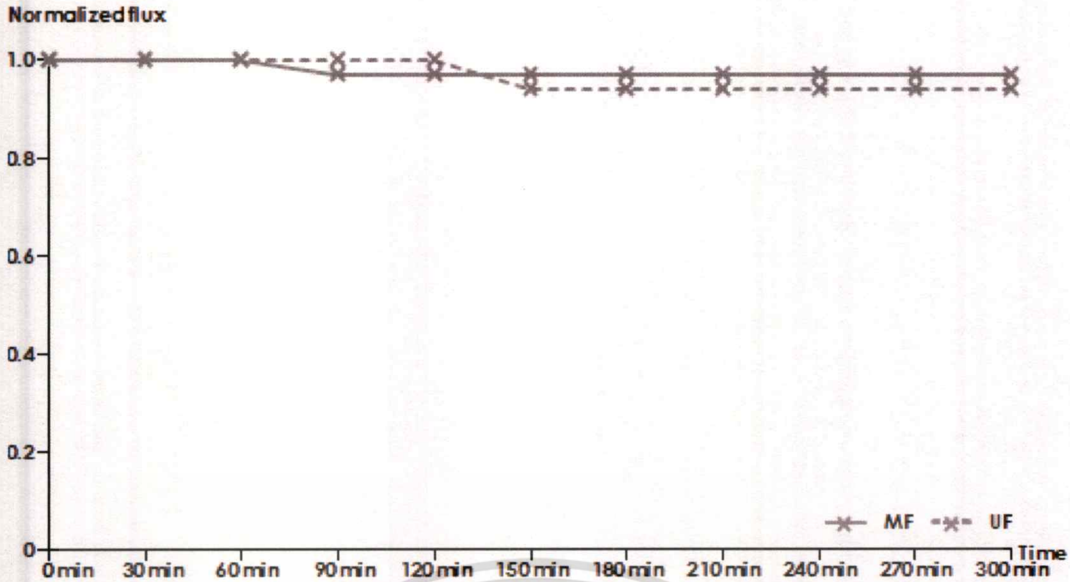


Figure 4.9 Normalized flux of MF and UF membranes at various times

4.2.2 Effect of chitosan:formaldehyde ratio on formaldehyde removal and flux decline by microfiltration (MF) and ultrafiltration (UF) membranes

Figure 4.10 illustrated percent removal based on formaldehyde concentration of MF and UF membranes at chitosan:formaldehyde ratio of 100:1, 200:1, and 300:1. The results showed that percent removal increased when chitosan:formaldehyde ratio increased. Overall efficiency in formaldehyde removal were in a range of 35-36%, 50-51%, and 59-60% at chitosan:formaldehyde ratio of 100:1, 200:1, and 300:1, respectively. It can be explained that amount of chitosan increased as chitosan:formaldehyde ratio increased that provided more active site to react with formaldehyde cause it higher percent removal of formaldehyde. When compare percent removal between MF and UF membranes, it was found that percent removal of these membranes was similar. It can be explained that formaldehyde was not retained by MF and UF membranes, thus almost formaldehyde was absorbed by chitosan. Then chitosan binding formaldehyde was retained by MF and UF membranes.

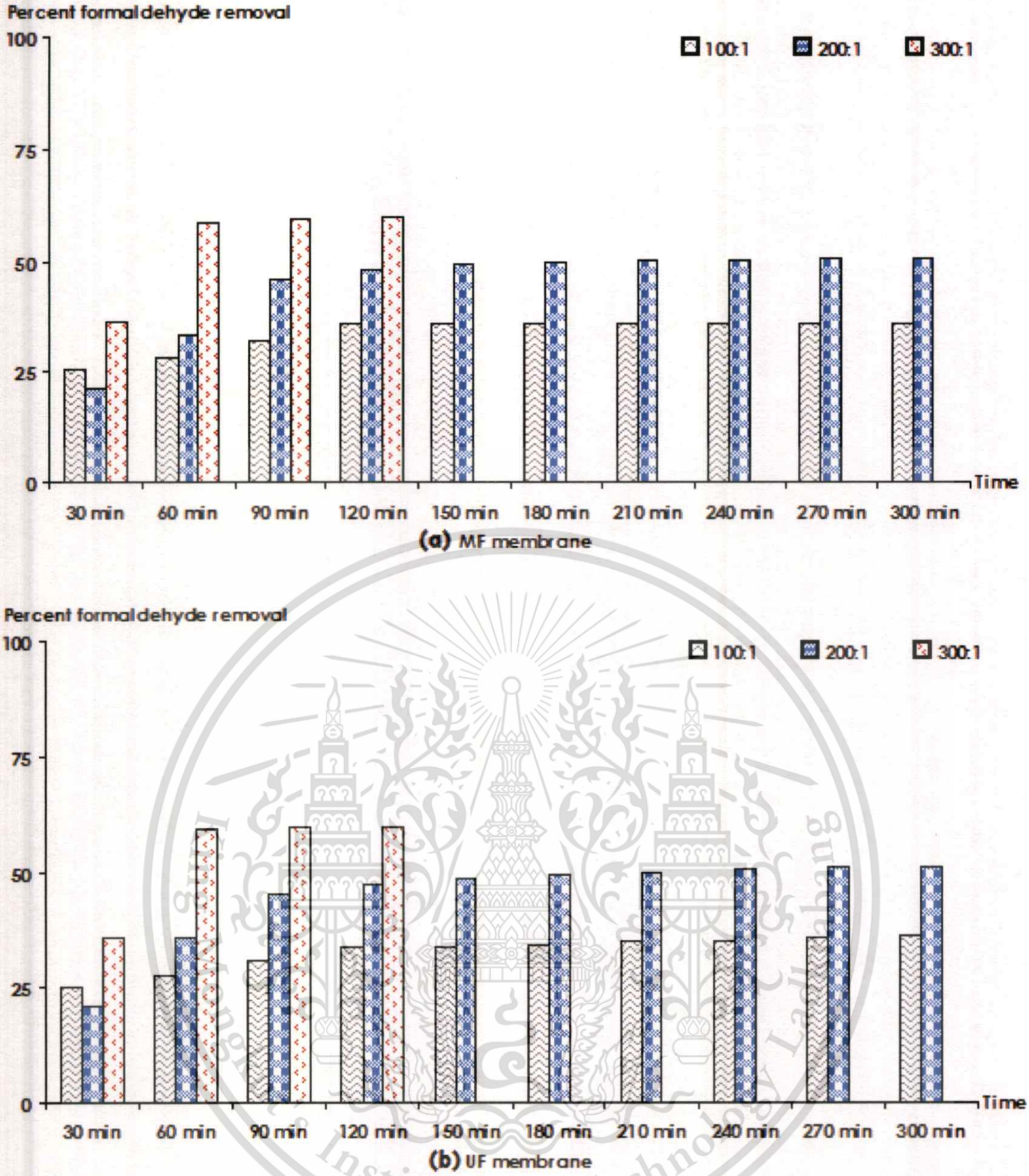


Figure 4.10 Percent removal based on formaldehyde concentration of (a) MF membrane and (b) UF membrane at chitosan:formaldehyde ratio of 100:1, 200:1, and 300:1

From the experiment, only chitosan:formaldehyde ratio 100:1, 200:1, and 300:1 were operated. At higher ratio, the membranes were found to be clogged due to a large amount of chitosan at feed inlet, causing no permeate flowing out from the system. At chitosan:formaldehyde of 300:1, the system was operated for 2 hours before the system was totally clogged. Chitosan flake accumulated and fouled membranes rapidly, leading to large membrane resistance untill no permeate was produced.

Figure 4.11 shows that normalized flux of permeate filtrated through MF and UF membranes decreased when increasing chitosan:formaldehyde ratio. It can be explained that an increase of chitosan:formaldehyde ratio caused amount of chitosan in the solution increased, the membrane were found to be clogged on membrane surface or in membrane pores due to solute from a large amount of chitosan in the solution and permeate flux of MF and UF membranes decreased.

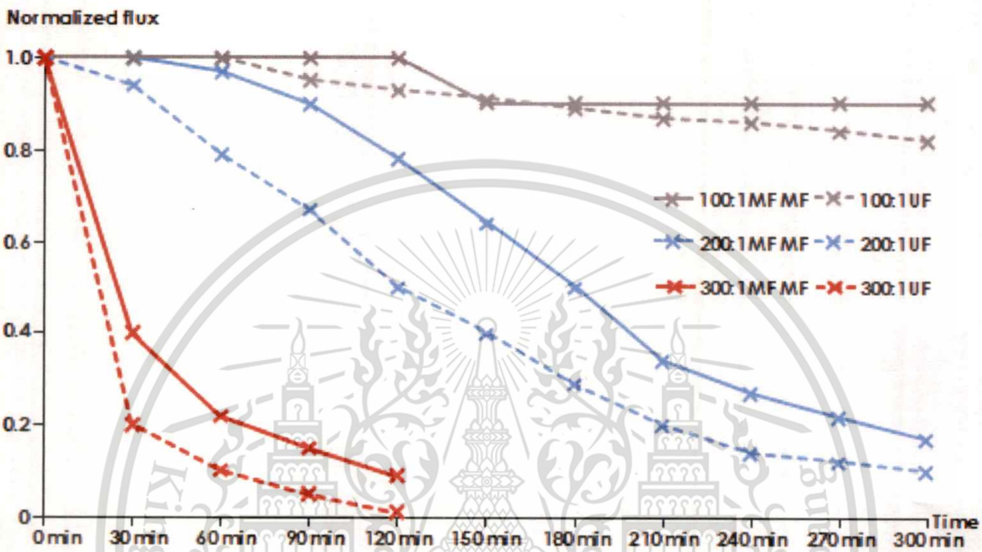


Figure 4.11 Normalized flux of MF and UF membranes at chitosan:formaldehyde ratio of 100:1, 200:1, and 300:1

Figure 4.12 showed percent flux decline of MF and UF membranes at 120 min of chitosan:formaldehyde ratio 100:1, 200:1, and 300:1, respectively. It was found that percent flux decline decreased as chitosan:formaldehyde increased. At chitosan:formaldehyde ratio of 300:1, exhibited highest flux decline compared to 100:1 and 200:1 ratio, which was not practical for membrane operation. When compared between MF and UF membranes at chitosan:formaldehyde ratio of 100:1, 200:1, and 300:1, it was found that percent flux decline of MF was less than UF membrane although formaldehyde removal percentage of MF and UF membranes were similar.

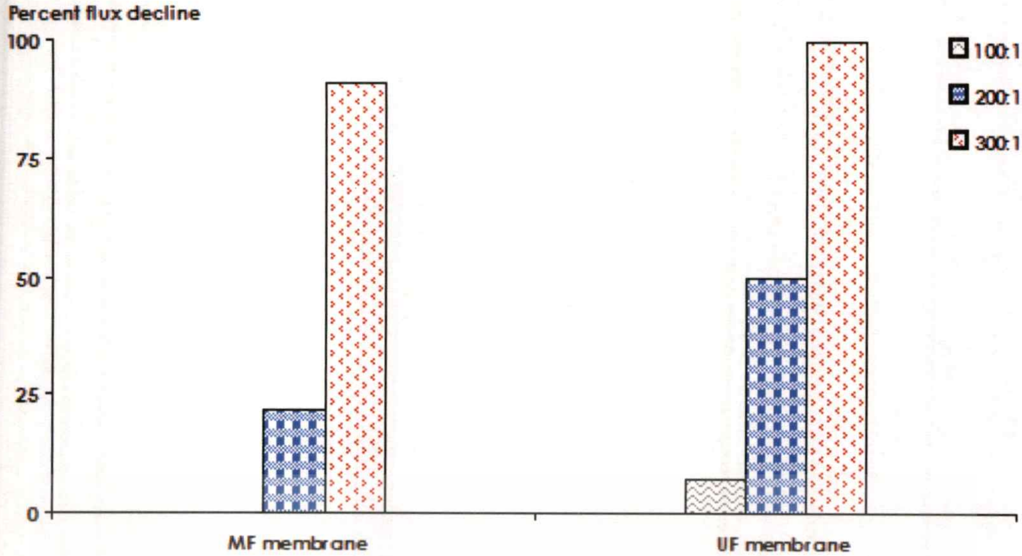


Figure 4.12 Percent flux decline of MF and UF membranes at 120 min of chitosan:formaldehyde ratio 100:1, 200:1, and 300:1

When considering DOC remaining in permeate from MF and UF membranes at chitosan:formaldehyde ratio of 100:1, 200:1, and 300:1, it was found that DOC remaining in MF permeate was higher than that from UF membrane as shown in Figure. 4.13. Organic carbon dissociated from chitosan could pass through MF membrane, whereas UF membrane could retain those organic compounds due to size exclusion effect. This phenomenon reflected the physical properties of permeate. As shown in Fig. 4.14, turbidity UF of permeate was less than that from MF.

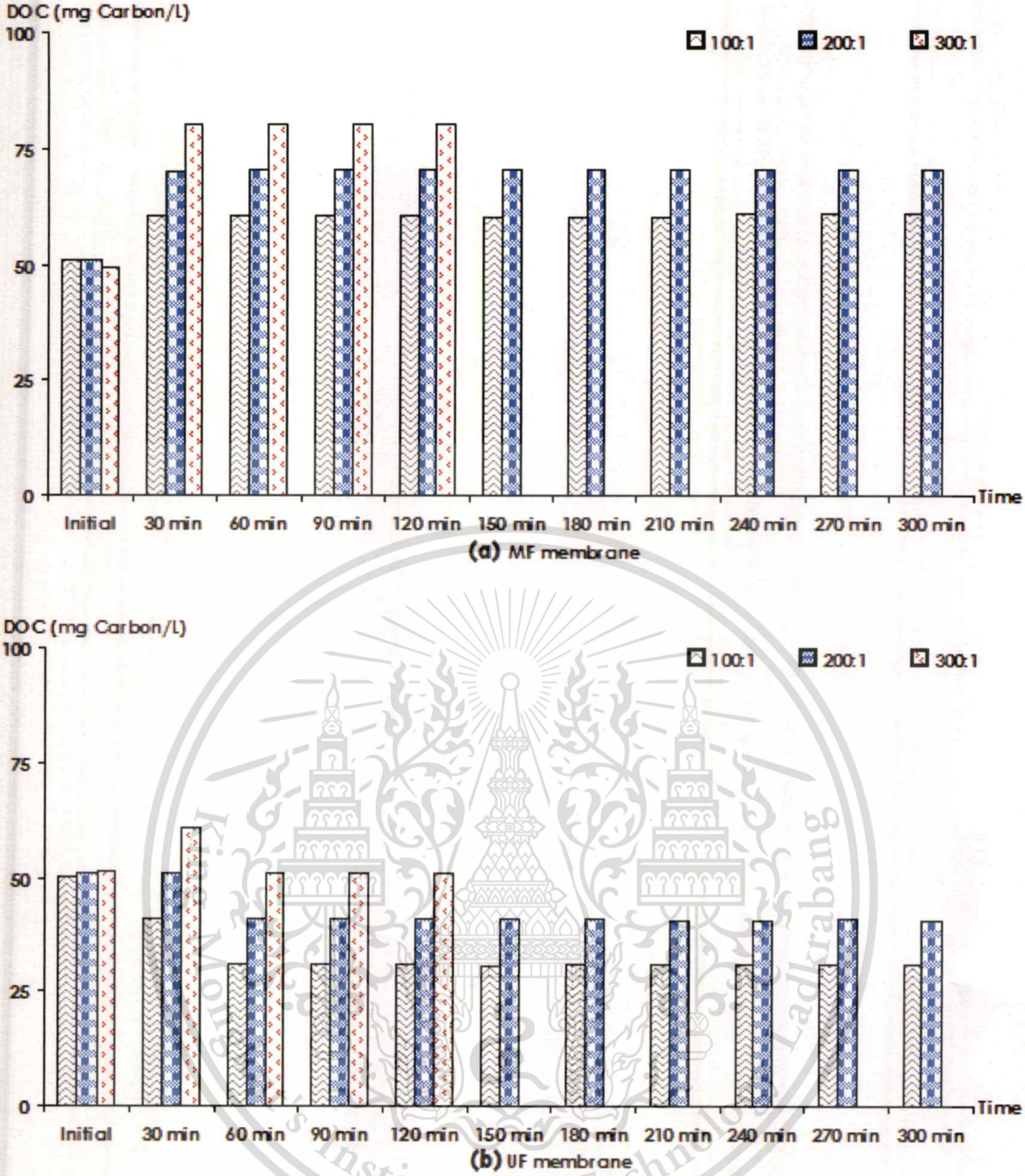


Figure 4.13 DOC remaining in the effluent of (a) MF membrane and (b) UF membrane at 100:1, 200:1, and 300:1 chitosan:formaldehyde ratio

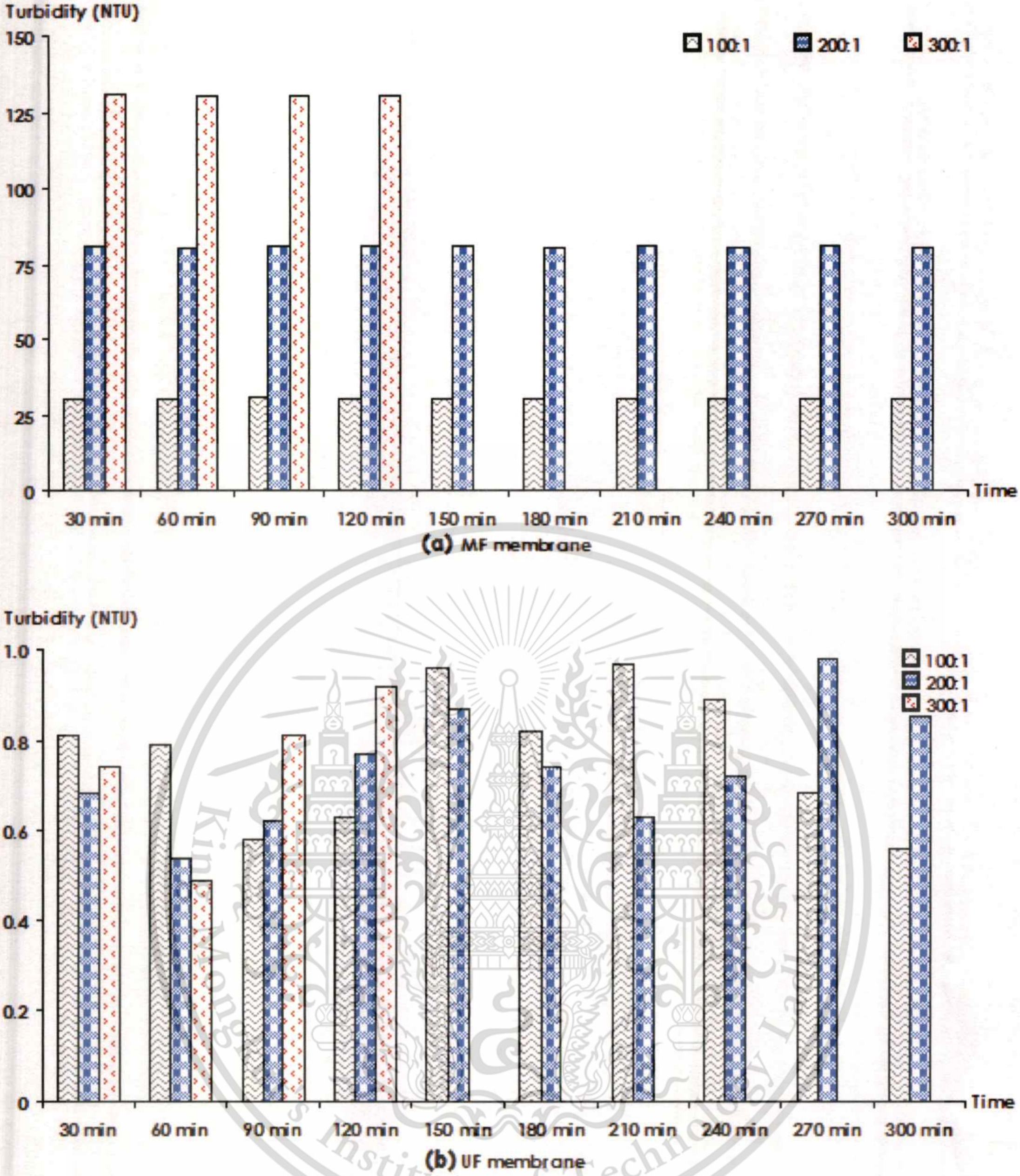


Figure 4.14 Turbidity remaining in the effluent of (a) MF membrane and (b) UF membrane at 100:1, 200:1, and 300:1 chitosan:formaldehyde ratio

4.2.3 Effect of chitosan size on formaldehyde removal and flux decline by microfiltration (MF) and ultrafiltration (UF) membranes

Figure 4.15 (a) and (b) show percent removal of formaldehyde using chitosan-enhanced MF and UF membrane filtration, respectively, for different size of chitosan. The results show that all size of chitosan flake provided similar percent removal in a range of 50-51%. It can be explained that almost formaldehyde was absorbed by amino groups in chitosan molecules and that a decrease in size of chitosan did not reduce amount of amino groups. Thus, chitosan flake size did not affect the removal efficiency of chitosan-enhanced membrane filtration. The results also exhibited that MF and UF membranes did not remove formaldehyde directly, but chitosan played significant role in adsorption of formaldehyde as chemisorption[27]. Then, the combined of chitosan-formaldehyde compounds were retained by membranes.

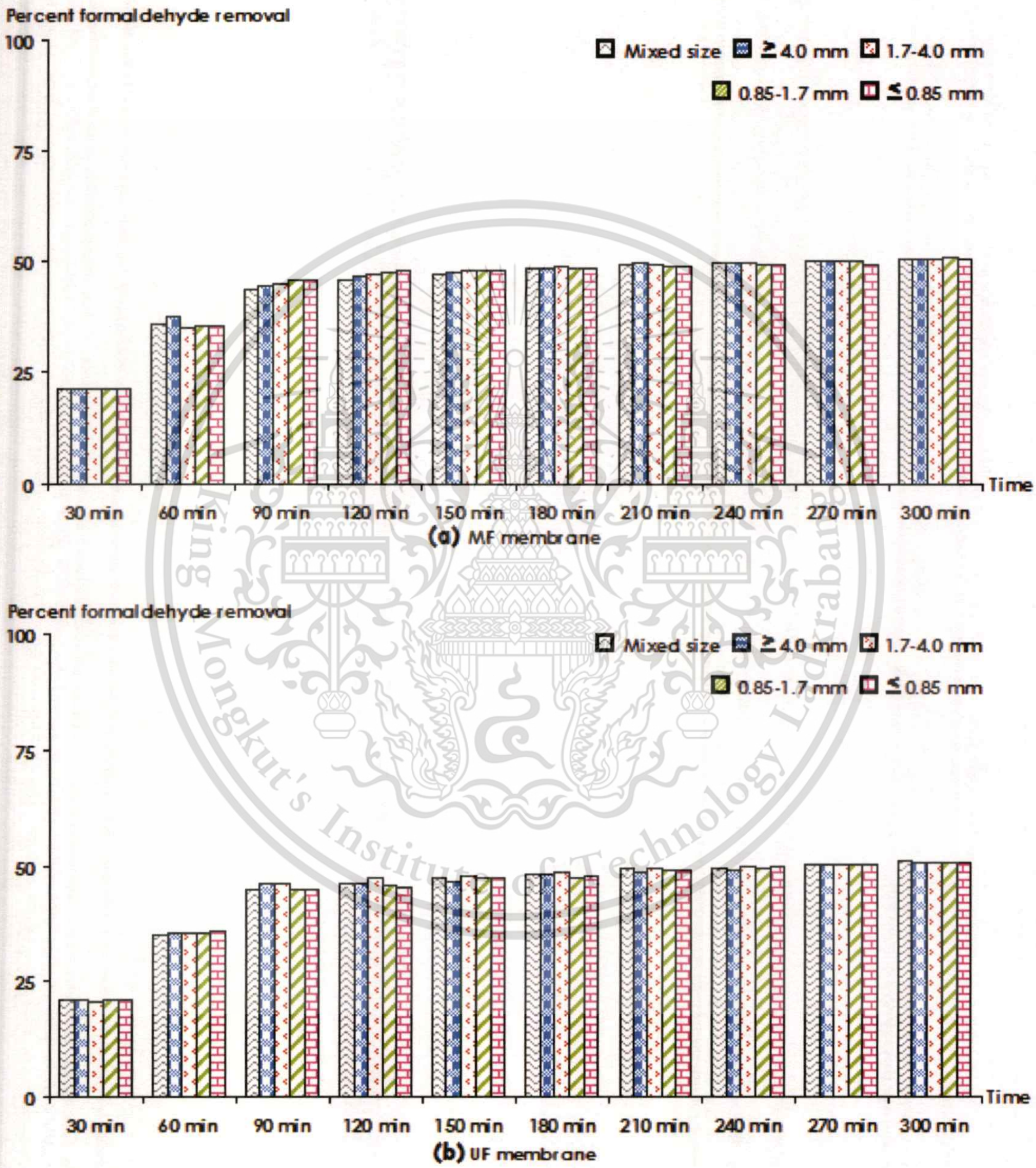


Figure 4.15 Percent removal based on formaldehyde concentration of (a) MF membrane and (b) UF membranes at different size of chitosan flake

Figure 4.16 illustrates percent flux decline at 120 min of MF and UF membranes at chitosan size of ≤ 0.85 mm, 0.85-1.7 mm, 1.7-4.0 mm, ≥ 4.0 mm and mixed size. The results showed that percent flux decline for all sizes of chitosan flake were similar for MF membrane but percent flux decline slightly decreased when size of chitosan decreased for UF membrane. It can be explained that chitosan in feed reservoir was retained by strainer of which pore size was smaller than chitosan flake (420 micron or 0.42 mm). Only solutes smaller than pores size passed through strainer and fouled membrane. The membranes were found to be clogged on membrane surface or in membrane pores and permeate flux of MF and UF membranes decreased.

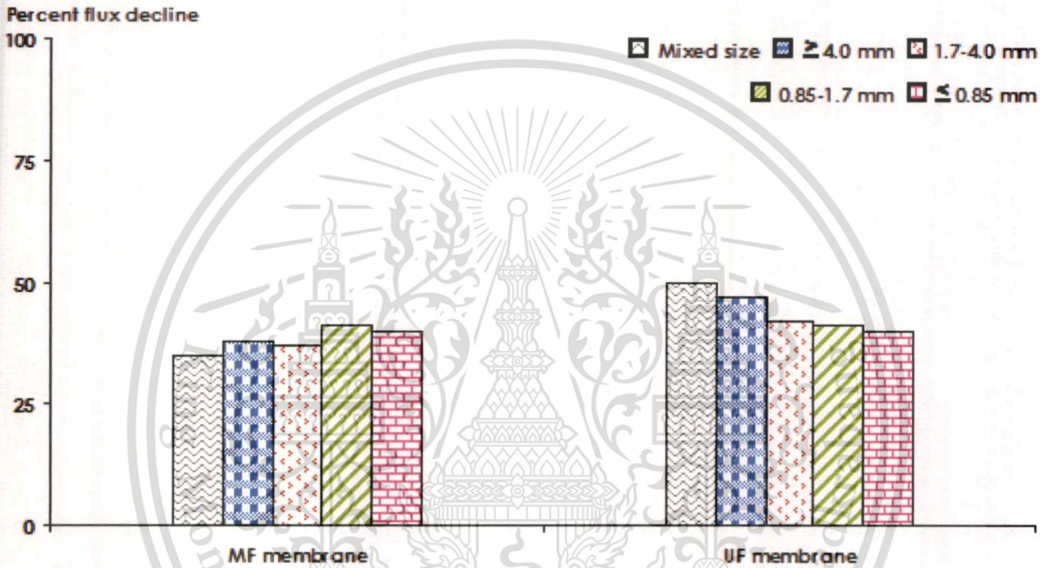


Figure 4.16 Percent flux decline of MF and UF membranes at different size of chitosan flake

Figure 4.17 showed that DOC remaining in permeate from UF membrane was less than that in MF permeate because UF membrane could retain organic carbon dissociated from chitosan better than MF membrane. Besides, UF membrane also retained turbidity of permeate more than MF membrane due to size exclusion effect. Smaller size of chitosan flake was more soluble because of higher surface area and provided a large amount of solutes in the solution. Solutes in the solution could pass through MF membrane because it was smaller than pore size of MF membrane, whereas it was retained by UF membrane due to pore size of this membrane was smaller than those solutes in the solution.

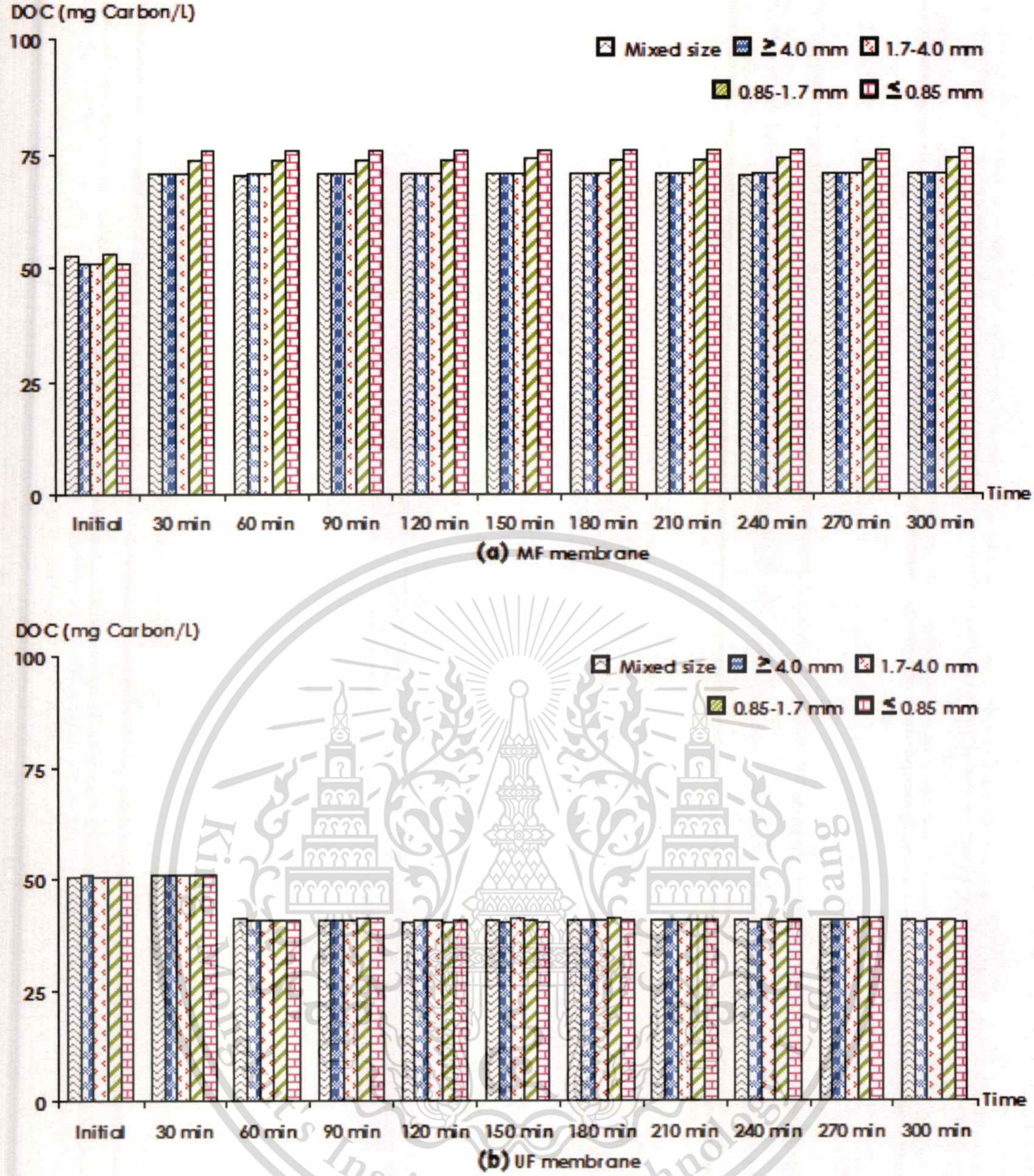


Figure 4.17 DOC remaining in the effluent of (a) MF membrane and (b) UF membrane at different size of chitosan flake

4.2.4 Effect of pH of formaldehyde solution on formaldehyde removal and flux decline by microfiltration (MF) and ultrafiltration (UF) membranes

In the experiment, formaldehyde synthetic wastewater at original pH and pH of 4, 7 and 10 was used to operate the system in order to study the effect of pH of formaldehyde solution on formaldehyde removal by MF and UF membranes. The results showed that pH of formaldehyde solution did not affect formaldehyde removal by MF and UF membranes and formaldehyde removal efficiency were similar. It can be explained that formaldehyde had molecular size less than pore size of membranes, thus formaldehyde molecules were not retained by MF and UF membranes. Figure 4.18 shows percent removal based on formaldehyde concentration of MF and UF membranes at original pH and pH of 4, 7 and 10. The results showed that all pH of formaldehyde solution provided similar percent removal in a range of 50-51%.

It can be explained that the reaction between chitosan and formaldehyde was crosslinked by formaldehyde as a crosslinking agent. When chitosan was mixed with formaldehyde solution, a part of chitosan that partial dissolved in the solution was crosslinked by formaldehyde between amino group of chitosan and oxygen atom of formaldehyde, while chitosan that did not dissolved in the solution reacted with formaldehyde and formed covalent bond. A variation in pH of solution did not affect crosslinking or covalent bond between chitosan and formaldehyde. Thus, percent removal of formaldehyde at different pH of solution was similar.

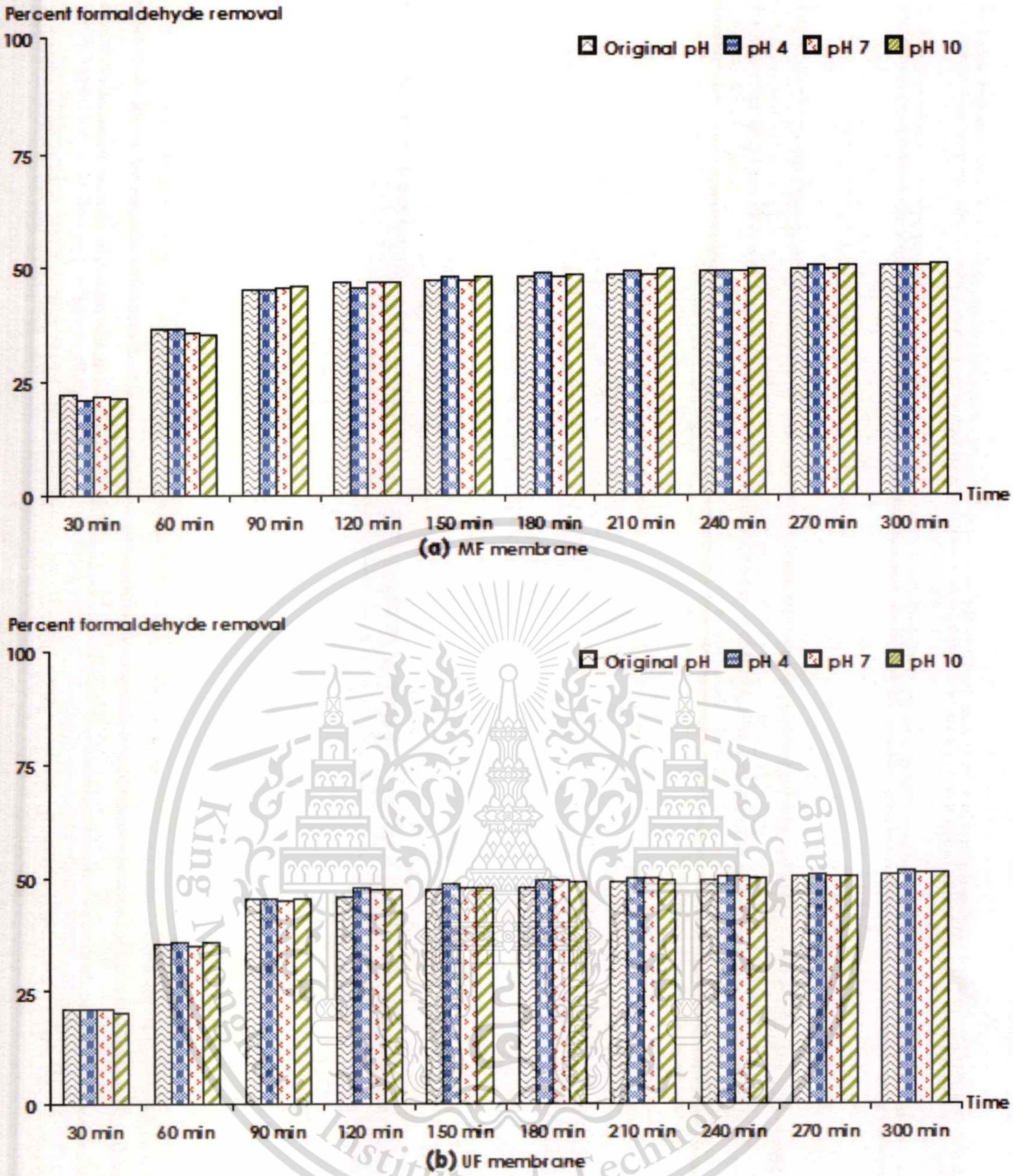


Figure 4.18 Percent removal based on formaldehyde concentration of (a) MF membrane and (b) UF membrane at different pH of formaldehyde solution

As illustrated in Fig 4.19, percent flux decline of MF and UF membranes at original pH and pH of 4, 7 and 10 were similar. It can be explained that pH of formaldehyde solution did not affect for the dissolution of chitosan and it did not increase solutes in the solution, thus percent flux decline at all pH range were similar.

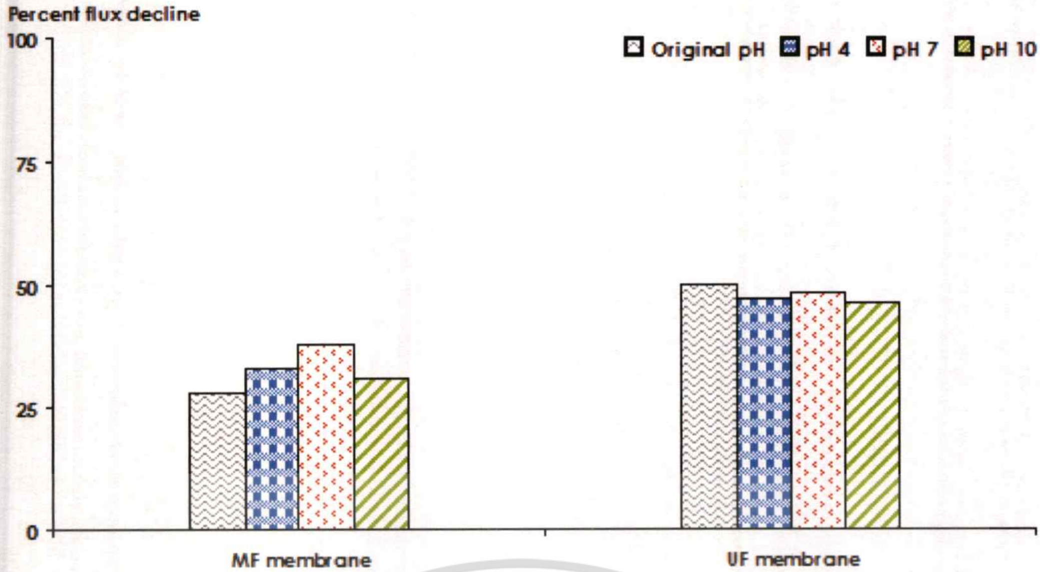


Figure 4.19 Percent flux decline of MF and UF membrane at different pH of formaldehyde solution

When considering DOC remaining in the permeate from MF and UF membranes at original pH and pH of 4, 7 and 10, it showed that UF membrane retained organic carbon in the solution more than MF membrane, as illustrated in Fig. 4.20. Besides, turbidity from the dissolution of chitosan was also retained by UF membranes more than MF membrane (data not shown).

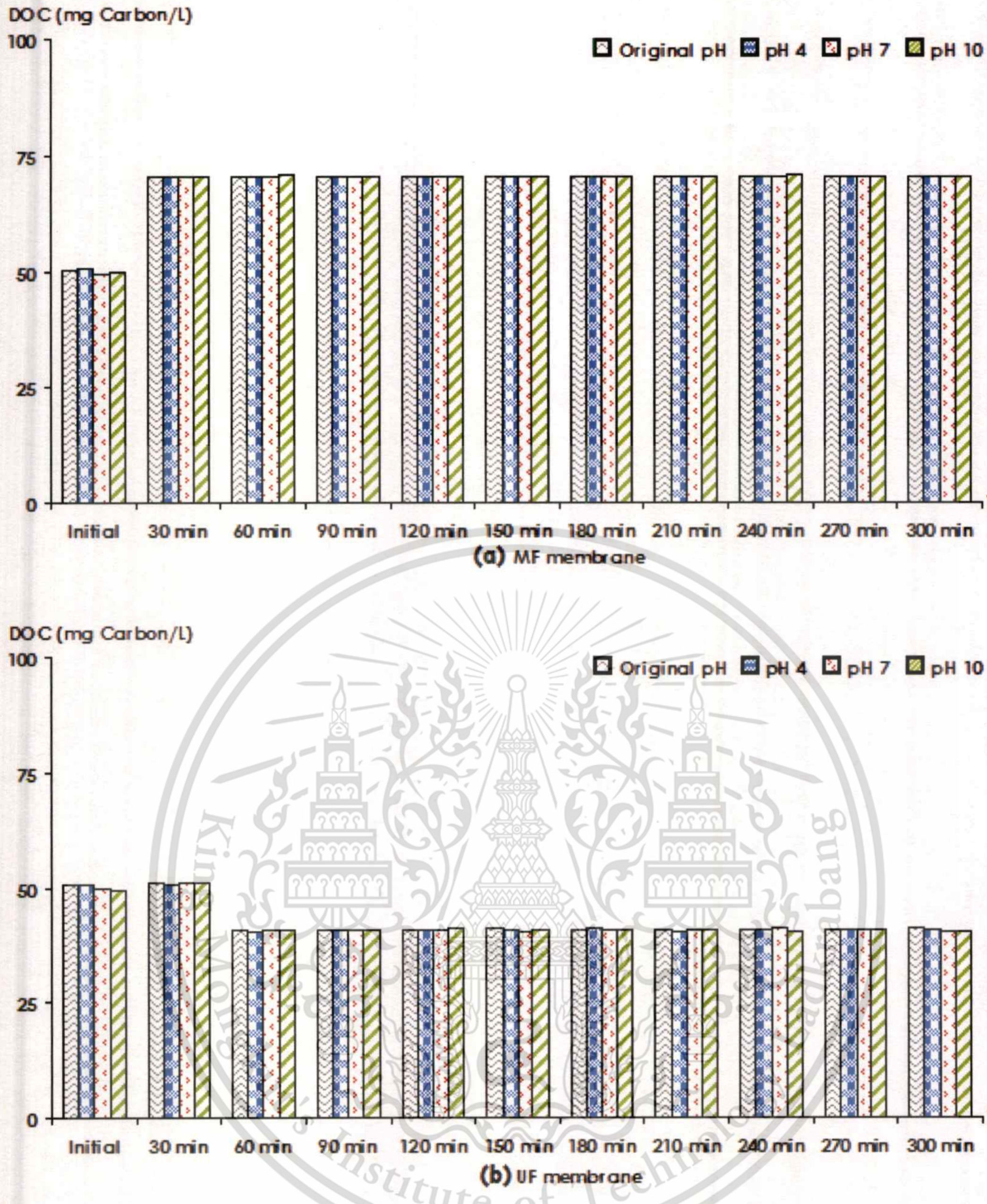


Figure 4.20 DOC remaining in the effluent of (a) MF membrane and (b) UF membrane at different pH of formaldehyde solution

4.3 Comparison of adsorption and membrane processes on formaldehyde, dissolved organic carbon, and turbidity removal

The performance of chitosan adsorption and chitosan-enhanced membrane process on formaldehyde, dissolved organic carbon, and turbidity removal were investigated. Similarly, the adsorption and membrane process were found to be efficiency in formaldehyde removal with removal efficient of 35-36%, 50-51%, and 59-60% at chitosan:formaldehyde ratio of 100:1, 200:1, and 300:1, respectively, as illustrated in Fig. 4.21.

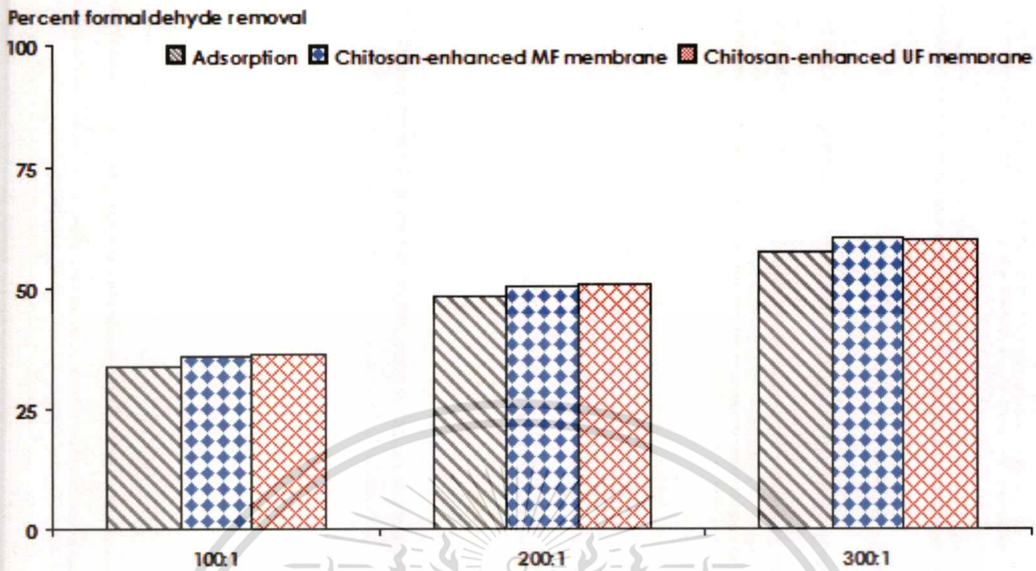


Figure 4.21 Comparison of formaldehyde removal efficiency between adsorption and membrane processes

DOC and turbidity generated from organic carbon and suspended particles from the dissolution of chitosan were monitored. In the adsorption and MF process, DOC still remained in the solution after adsorption and MF process while organic carbon was retained by UF membrane. The comparison of DOC between adsorption process and membrane process was shown in Fig. 4.22 (a). Chitosan-enhanced membrane process exhibited higher efficiency in terms of DOC and turbidity removal. Turbidity in adsorption process was less than that in MF process because, in adsorption process, chitosan and formaldehyde synthetic wastewater was mixed by orbital shaker while it was stirred directly by agitator in membrane process that increased the dissolution of chitosan. Besides, in analysis of turbidity from the adsorption process, formaldehyde mixed with chitosan was filtered through 0.45-μm filter paper which employed smaller pore size than MF membrane. Thus, turbidity in adsorption process was less than in MF membrane, as illustrated in Fig. 4.22 (b).

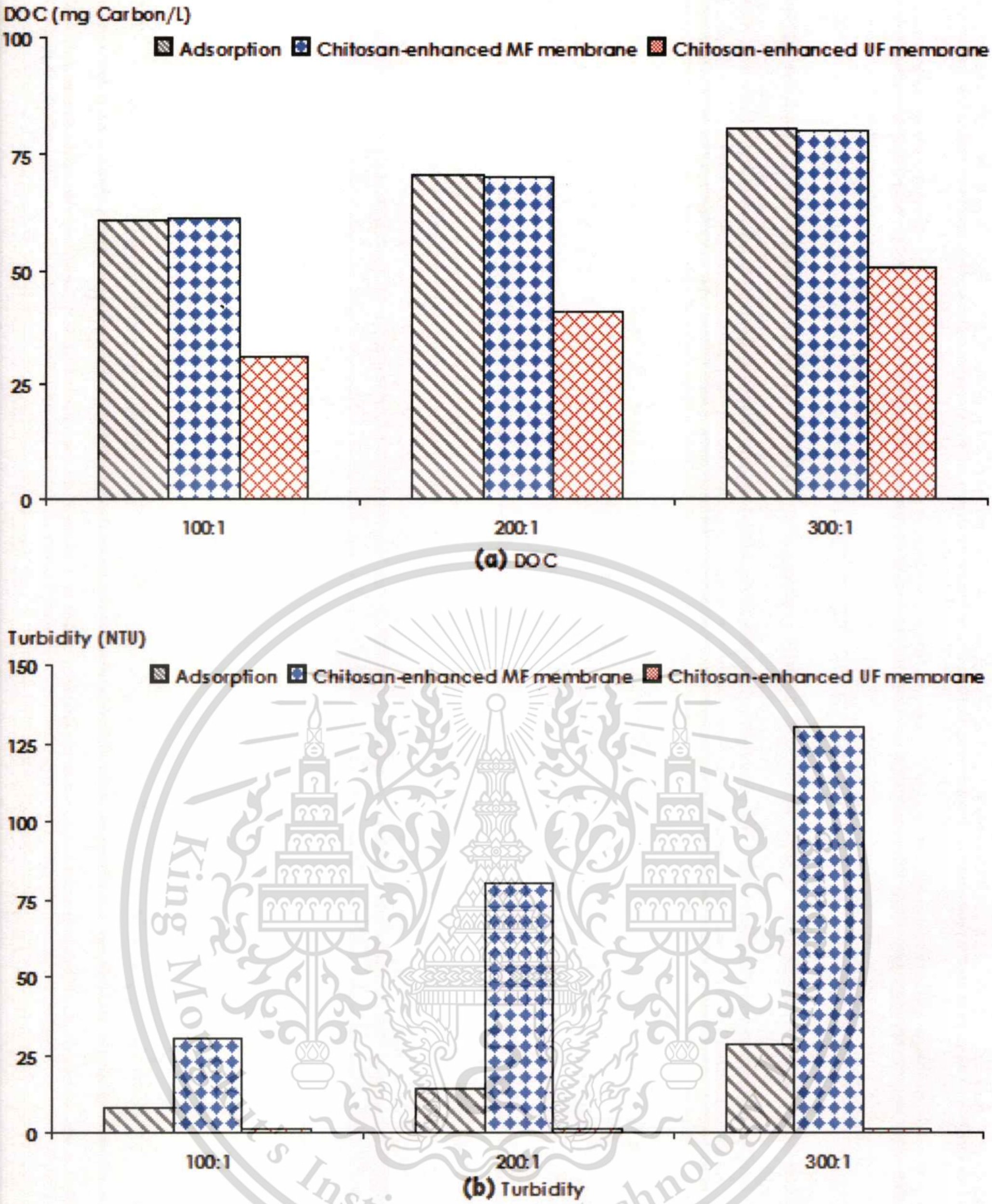


Figure 4.22 Comparison of (a) DOC and (b) turbidity remaining in the solution between adsorption and membrane processes

When compared removal efficiency in terms of DOC and turbidity between MF and UF membranes, it was shown that MF membrane did not remove DOC but partially removed turbidity while UF partially removed DOC and totally removed turbidity. The effluent from UF membrane appeared transparent without turbidity as shown in Fig. 4.23.

(a) (b)

Figure 4.23 Appearance of effluent between (a) MF membrane and (b) UF membrane

CHAPTER 5

CONCLUSIONS AND SUGGESTIONS

5.1 Conclusions

- Chitosan adsorption test

In chitosan adsorption test, chitosan flake with 85% deacetylation and molecular weight 50,000 daltons was used as absorbent. Increasing chitosan:formaldehyde ratio increased percent removal, however, DOC and turbidity in the solution also increased. A decrease in size of chitosan flake did not affect formaldehyde removal, but smaller size of chitosan flake caused high DOC and turbidity levels in the solution. The pH of formaldehyde solution and mixing time more than 0.5 hours did not exhibited any effect on formaldehyde adsorption, DOC, and turbidity. With initial formaldehyde concentration of 120 mg/L at original pH and chitosan:formaldehyde ratio of 500:1, it was found that chitosan was able to remove formaldehyde up to 71-72% at mixing time 0.5 hours.

- Chitosan-enhanced membrane filtration process

In chitosan-enhanced membrane process, increasing chitosan:formaldehyde ratio increased percent formaldehyde removal, DOC, and turbidity in the effluent. Besides, membranes were found to be clogged due to a large amount of chitosan when increasing chitosan:formaldehyde ratio and percent flux decline also decreased. Chitosan size and pH of formaldehyde did not exhibited effect on formaldehyde removal, whereas DOC and turbidity in the effluent slightly increased when chitosan size decreased. When compared percent flux decline of MF and UF membranes, it was found that decreasing of chitosan size was slightly decrease percent flux decline of UF membranes due to size exclusion effect.

- Comparison of chitosan adsorption and chitosan-enhanced membrane filtration process

Adsorption by chitosan and chitosan-enhanced membrane filtration exhibited a potential in formaldehyde removal. Membrane process exhibited higher efficiency than adsorption in terms of DOC and turbidity removal. When compared between MF and UF membranes, UF membrane exhibited higher efficiency than MF membrane in terms of DOC and turbidity removal. MF membrane did not remove DOC but partially removed turbidity while UF partially removed DOC and totally removed turbidity.

5.2 Suggestions

1) To improve percent removal of formaldehyde, chitosan with higher % deacetylation should be investigated.

2) For better quality of effluent, integrated membrane system using MF followed by UF membrane should be investigated for removal DOC and turbidity remaining in the effluent.

3) To reduce a potential of flux decline during membrane filtration, other types of membranes, e.g. tubular, and plate and frame membrane, should be investigated.

4) Due to the partial dissolution of chitosan flake, the ratio of active chitosan and formaldehyde could not be controlled. Therefore, other chitosan forms, e.g. solution, beads, or modified-chitosan, should be investigated.

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APPENDIX

EXPERIMENTAL DATA

EXPERIMENT A

ADSORPTION PROCESS

Table A.1 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH of solution by adsorption process to study mixing time as preliminary test

	FM Conc. (mg/L)			%Removal			DOC (mg Carbon/L)			Turbidity (NTU)			pH		
	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.
Initial FM	125.88	125.30	125.59				50.87	50.37	50.62	0.23	0.25	0.24	7.85	7.86	7.86
Initial FM 12 Hrs (Control)	125.20	124.80	125.00				50.65	50.59	50.62	0.25	0.25	0.25	7.83	7.84	7.84
Initial FM 24 Hrs (Control)	124.53	124.31	124.42				50.43	50.67	50.55	0.25	0.24	0.25	7.81	7.83	7.82
Mixing time 12 Hrs	35.77	35.38	35.58	71.43	71.65	71.54	100.43	100.72	100.58	43.7	43.1	43.4	8.54	8.55	8.55
Mixing time 24 Hrs	34.42	34.39	34.41	72.36	72.34	72.35	100.79	100.44	100.62	45.8	45.3	45.6	8.56	8.58	8.57

Table A.2 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH of solution by adsorption process to study the effect of chitosan:formaldehyde ratio

	FM Conc. (mg/L)			%Removal			DOC (mg Carbon/L)			Turbidity (NTU)			pH		
	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.
Initial FM	125.72	123.44	124.58				50.87	50.77	50.82	0.27	0.30	0.29	7.87	7.86	7.87
Initial FM (Control)	125.34	123.06	124.20				50.54	50.51	50.52	0.25	0.29	0.27	7.84	7.81	7.83
100:1 CS:FM ratio	83.11	82.34	82.72	33.70	33.09	33.39	60.78	60.64	60.71	7.76	7.75	7.76	7.95	7.93	7.94
200:1 CS:FM ratio	64.84	63.32	64.08	48.27	48.55	48.41	70.39	70.81	70.60	14.5	15.1	14.8	8.07	8.07	8.07
300:1 CS:FM ratio	53.42	52.28	52.85	57.38	57.51	57.44	80.61	80.92	80.76	28.5	28.7	28.6	8.15	8.15	8.15
400:1 CS:FM ratio	45.81	45.05	45.43	63.45	63.39	63.42	90.54	90.53	90.53	38.5	40.4	39.5	8.38	8.39	8.39
500:1 CS:FM ratio	34.40	33.64	34.02	72.56	72.67	72.61	100.37	100.77	100.57	45.8	48.1	47.0	8.59	8.55	8.57

Table A.3 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH of solution by adsorption process to study the effect of chitosan size

	FM Conc. (mg/L)			%Removal			DOC (mg Carbon/L)			Turbidity (NTU)			pH		
	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.
Initial FM	125.75	123.98	124.86				50.91	50.32	50.61	0.25	0.28	0.27	7.88	7.86	7.87
Initial FM (Control)	125.31	123.53	124.42				50.67	50.87	50.77	0.23	0.23	0.23	7.85	7.82	7.83
Mixed size chitosan	34.72	34.28	34.50	72.29	72.25	72.27	100.54	100.38	100.46	45.8	45.7	45.8	8.58	8.57	8.57
Chitosan size ≥ 4.0 mm	35.61	35.61	35.61	71.58	71.17	71.37	100.82	100.13	100.47	45.2	44.2	44.7	8.55	8.53	8.54
Chitosan size 1.7-4.0 mm	35.61	35.61	35.61	71.58	71.17	71.37	100.69	100.56	100.62	45.9	45.1	45.5	8.57	8.55	8.56
Chitosan size 0.85-1.7 mm	36.06	35.17	35.62	71.23	71.53	71.38	104.93	105.74	105.33	50.8	51.9	51.4	8.60	8.59	8.59
Chitosan size ≤ 0.85 mm	35.17	34.72	34.94	71.93	71.89	71.64	110.78	110.89	110.83	55.3	53.8	54.6	8.61	8.61	8.61

Table A.4 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH of solution by adsorption process to study the effect of pH of formaldehyde solution

	FM Conc. (mg/L)			%Removal			DOC (mg Carbon/L)			Turbidity(NTU)			pH		
	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.
Initial FM - Original pH	115.71	115.29	115.50				48.61	48.59	48.60	0.30	0.32	0.31	7.85	7.86	7.85
Initial FM - pH 4	114.86	114.44	114.65				48.55	48.29	48.42	0.31	0.31	0.31	4.05	4.05	4.05
Initial FM - pH 7	114.02	114.86	114.44				48.73	48.78	48.75	0.28	0.30	0.29	7.07	7.05	7.06
Initial FM - pH 10	114.44	114.02	114.65				48.82	48.92	48.78	0.30	0.30	0.30	10.03	10.06	10.04
Initial FM - Original pH (Control)	115.29	114.86	115.07				48.59	48.50	48.54	0.30	0.30	0.30	7.83	7.82	7.82
Initial FM - pH 4 (Control)	114.44	114.44	114.44				48.77	48.26	48.51	0.30	0.28	0.29	4.03	4.02	4.02
Initial FM - pH 7 (Control)	113.60	114.02	113.81				48.64	48.56	48.60	0.29	0.31	0.30	7.04	7.01	7.02
Initial FM - pH 10 (Control)	114.02	113.60	113.81				48.33	48.92	48.62	0.29	0.31	0.30	10.01	10.03	10.02
Original pH	31.25	31.67	31.46	72.89	72.43	72.66	100.56	100.79	100.68	47.1	45.2	46.2	8.56	8.53	8.54
pH 4	32.09	32.52	32.30	71.96	71.59	71.77	100.71	100.61	100.66	45.8	45.9	45.9	4.74	4.75	4.74
pH 7	32.09	32.09	32.09	71.75	71.85	71.80	100.92	100.82	100.87	45.3	45.8	45.6	7.75	7.73	7.74
pH 10	32.09	32.09	32.09	71.85	71.75	71.80	100.84	100.93	100.89	45.1	45.4	45.3	9.23	9.22	9.22

Table A.5 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH of solution by adsorption process to study the effect of mixing time

	FM Conc. (mg/L)			%Removal			DOC (mg Carbon/L)			Turbidity(NTU)			pH		
	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.	#1	#2	Avr.
Initial FM	118.90	125.87	122.38				50.39	50.39	50.39	0.33	0.31	0.32	7.88	7.89	7.88
Initial FM 0.25 Hrs (Control)	118.03	125.87	121.95				50.23	50.33	50.28	0.30	0.28	0.29	7.84	7.87	7.85
Initial FM 0.5 Hrs (Control)	118.03	125.87	121.95				50.11	50.39	50.25	0.28	0.30	0.29	7.85	7.83	7.84
Initial FM 0.75 Hrs (Control)	118.03	125.87	121.95				50.95	50.30	50.62	0.34	0.32	0.33	7.83	7.84	7.83
Initial FM 1 Hrs (Control)	118.03	125.44	121.73				50.54	50.22	50.38	0.29	0.30	0.29	7.86	7.87	7.86
Initial FM 2 Hrs (Control)	118.03	125.44	121.73				50.96	50.25	50.61	0.31	0.28	0.29	7.84	7.83	7.83
Initial FM 4 Hrs (Control)	117.60	125.44	121.52				50.10	50.30	50.20	0.30	0.31	0.30	7.81	7.85	7.83
Initial FM 8 Hrs (Control)	117.16	125.00	121.08				50.20	50.21	50.20	0.29	0.29	0.29	7.87	7.84	7.85
Initial FM 12 Hrs (Control)	116.72	124.56	120.64				50.33	50.13	50.23	0.31	0.33	0.32	7.83	7.86	7.84
Mixing time 0.25 Hrs	37.89	36.15	37.02	67.90	71.28	69.59	100.95	100.58	100.76	45.3	47.5	46.4	8.56	8.57	8.56
Mixing time 0.5 Hrs	33.97	35.71	34.84	71.22	71.63	71.42	100.85	100.65	100.75	47.1	47.2	47.2	8.58	8.53	8.55
Mixing time 0.75 Hrs	33.54	35.28	34.41	71.59	71.97	71.78	100.73	100.62	100.67	47.7	47.9	47.8	8.57	8.57	8.57
Mixing time 1 Hrs	33.10	34.84	33.97	71.96	72.22	72.09	100.87	100.79	100.83	47.3	47.6	47.5	8.59	8.55	8.57
Mixing time 2 Hrs	32.67	34.41	33.54	72.32	72.57	72.44	100.64	100.55	100.59	47.9	47.2	47.6	8.54	8.56	8.55
Mixing time 4 Hrs	32.67	34.41	33.54	72.22	72.57	72.44	100.71	100.67	100.69	47.4	47.3	47.4	8.55	8.58	8.56
Mixing time 8 Hrs	32.23	34.41	33.32	72.49	72.47	72.48	100.54	100.58	100.56	47.1	47.9	47.5	8.58	8.54	8.56
Mixing time 12 Hrs	31.79	33.97	32.88	72.76	72.73	72.74	100.68	100.98	100.83	47.2	47.6	47.4	8.56	8.55	8.55

EXPERIMENT B

CHITOSAN-ENHANCED MF MEMBRANE FILTRATION

Table B.1 Permeate flux of formaldehyde solution by microfiltration membrane at various times

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.18	2.18	1.00
5 min	2.18	2.18	1.00
10 min	2.18	2.18	1.00
15 min	2.18	2.18	1.00
30 min	2.18	2.18	1.00
45 min	2.18	2.18	1.00
60 min	2.18	2.18	1.00
75 min	2.18	2.12	0.97
90 min	2.18	2.12	0.97
105 min	2.18	2.12	0.97
120 min	2.18	2.12	0.97
135 min	2.18	2.12	0.97
150 min	2.18	2.12	0.97
165 min	2.18	2.12	0.97
180 min	2.18	2.12	0.97
195 min	2.18	2.12	0.97
210 min	2.18	2.12	0.97
225 min	2.18	2.12	0.97
240 min	2.18	2.12	0.97
255 min	2.18	2.12	0.97
270 min	2.18	2.12	0.97
285 min	2.18	2.12	0.97
300 min	2.18	2.12	0.97

Table B.2 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by microfiltration membrane at various times

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.53		50.15	0.35	7.80
Initial FM 30 min	125.53		50.23	0.29	7.85
Initial FM 60 min	125.53		50.34	0.21	7.87
Initial FM 90 min	125.53		50.44	0.24	7.84
Initial FM 120 min	125.53		50.24	0.19	7.83
Initial FM 150 min	125.09		50.54	0.23	7.86
Initial FM 180 min	125.09		50.25	0.26	7.87
Initial FM 210 min	125.09		50.47	0.22	7.85
Initial FM 240 min	125.09		50.55	0.28	7.84
Initial FM 270 min	125.09		50.62	0.30	7.86
Initial FM 300 min	125.09		50.46	0.19	7.87
Permeate 30 min	125.09	0.35	50.23	0.33	7.85
Permeate 60 min	125.09	0.35	50.07	0.28	7.83
Permeate 90 min	125.09	0.35	50.87	0.32	7.84
Permeate 120 min	125.09	0.35	50.93	0.17	7.86
Permeate 150 min	124.65	0.71	50.55	0.14	7.82
Permeate 180 min	124.65	0.71	50.69	0.15	7.88
Permeate 210 min	124.65	0.71	50.32	0.22	7.87
Permeate 240 min	124.65	0.71	50.12	0.26	7.85
Permeate 270 min	124.65	0.71	50.11	0.30	7.82
Permeate 300 min	124.65	0.71	50.94	0.21	7.87

Table B.3 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at chitosan:formaldehyde ratio of 100:1

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.18	2.18	1.00
5 min	2.18	2.18	1.00
10 min	2.18	2.18	1.00
15 min	2.18	2.18	1.00
30 min	2.18	2.18	1.00
45 min	2.18	2.18	1.00
60 min	2.18	2.18	1.00
75 min	2.18	2.07	0.95
90 min	2.18	2.07	0.95
105 min	2.18	2.07	0.95
120 min	2.18	2.07	0.95
135 min	2.18	1.96	0.90
150 min	2.18	1.96	0.90
165 min	2.18	1.96	0.90
180 min	2.18	1.96	0.90
195 min	2.18	1.96	0.90
210 min	2.18	1.96	0.90
225 min	2.18	1.96	0.90
240 min	2.18	1.96	0.90
255 min	2.18	1.96	0.90
270 min	2.18	1.96	0.90
285 min	2.18	1.96	0.90
300 min	2.18	1.96	0.90

Table B.4 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at chitosan:formaldehyde ratio of 100:1

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	126.26		51.10	0.21	7.87
Initial FM 30 min	97.37		60.84	30.5	7.98
Initial FM 60 min	92.12		60.37	30.7	7.95
Initial FM 90 min	86.87		60.75	30.8	7.97
Initial FM 120 min	81.62		60.18	30.6	7.93
Initial FM 150 min	81.62		60.85	30.9	7.94
Initial FM 180 min	81.62		60.03	30.5	7.96
Initial FM 210 min	81.62		60.04	30.7	7.98
Initial FM 240 min	81.62		60.42	30.6	7.95
Initial FM 270 min	81.62		60.56	30.3	7.97
Initial FM 300 min	81.62		60.58	30.5	7.94
Permeate 30 min	94.22	25.37	60.59	30.7	7.98
Permeate 60 min	90.55	28.29	60.45	30.4	7.93
Permeate 90 min	85.82	32.03	60.54	30.9	7.97
Permeate 120 min	81.09	35.77	60.61	30.8	7.97
Permeate 150 min	81.09	35.77	60.07	30.6	7.94
Permeate 180 min	81.09	35.77	60.26	30.2	7.99
Permeate 210 min	81.09	35.77	60.10	30.5	7.97
Permeate 240 min	81.09	35.77	60.74	30.7	7.95
Permeate 270 min	81.09	35.77	60.75	30.6	7.93
Permeate 300 min	81.09	35.77	60.89	30.2	7.96

Table B.5 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at chitosan:formaldehyde ratio of 200:1

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.18	2.18	1.00
5 min	2.18	2.18	1.00
10 min	2.18	2.18	1.00
15 min	2.18	2.18	1.00
30 min	2.18	2.18	1.00
45 min	2.18	2.18	1.00
60 min	2.18	2.12	0.97
75 min	2.18	2.07	0.95
90 min	2.18	1.96	0.90
105 min	2.18	1.86	0.85
120 min	2.18	1.70	0.78
135 min	2.18	1.53	0.70
150 min	2.18	1.39	0.64
165 min	2.18	1.27	0.58
180 min	2.18	1.09	0.50
195 min	2.18	0.91	0.42
210 min	2.18	0.74	0.34
225 min	2.18	0.64	0.29
240 min	2.18	0.59	0.27
255 min	2.18	0.53	0.24
270 min	2.18	0.48	0.22
285 min	2.18	0.42	0.19
300 min	2.18	0.36	0.17

Table B.6 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at chitosan:formaldehyde ratio of 200:1

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	128.78		51.24	0.19	7.84
Initial FM 30 min	106.49		70.52	80.7	8.07
Initial FM 60 min	88.08		70.26	80.1	8.03
Initial FM 90 min	70.64		70.19	80.6	8.05
Initial FM 120 min	67.73		70.42	80.4	8.08
Initial FM 150 min	65.79		70.37	80.6	8.04
Initial FM 180 min	65.31		70.28	80.3	8.06
Initial FM 210 min	64.83		70.31	80.5	8.09
Initial FM 240 min	64.83		70.23	80.2	8.03
Initial FM 270 min	64.34		70.37	80.7	8.07
Initial FM 300 min	64.34		70.55	80.4	8.04
Permeate 30 min	101.65	21.07	70.21	80.9	8.08
Permeate 60 min	85.66	33.48	70.28	80.5	8.05
Permeate 90 min	69.67	45.90	70.33	80.8	8.06
Permeate 120 min	67.25	47.78	70.41	80.6	8.09
Permeate 150 min	65.31	49.29	70.24	80.9	8.03
Permeate 180 min	64.83	49.66	70.38	80.2	8.07
Permeate 210 min	64.34	50.04	70.44	80.8	8.05
Permeate 240 min	64.34	50.04	70.57	80.5	8.09
Permeate 270 min	63.86	50.41	70.65	80.7	8.06
Permeate 300 min	63.86	50.41	70.32	80.4	8.08

Table B.7 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at chitosan:formaldehyde ratio of 300:1

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.18	2.18	1.00
5 min	2.18	2.01	0.92
10 min	2.18	1.78	0.81
15 min	2.18	1.50	0.69
30 min	2.18	0.88	0.40
45 min	2.18	0.64	0.29
60 min	2.18	0.47	0.22
75 min	2.18	0.42	0.19
90 min	2.18	0.32	0.15
105 min	2.18	0.23	0.11
120 min	2.18	0.20	0.09

Table B.8 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at chitosan:formaldehyde ratio of 300:1

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	120.24		49.50	0.20	7.85
Initial FM 30 min	80.56		80.31	130.5	8.17
Initial FM 60 min	50.79		80.27	130.4	8.14
Initial FM 90 min	49.31		80.33	130.9	8.16
Initial FM 120 min	48.31		80.25	130.7	8.19
Permeate 30 min	76.59	36.30	80.35	130.8	8.13
Permeate 60 min	49.31	58.99	80.21	130.5	8.18
Permeate 90 min	48.81	59.41	80.20	130.4	8.19
Permeate 120 min	47.82	60.23	80.23	130.7	8.15

Table B.9 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at mixed size of chitosan

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.25	2.18	1.00
5 min	2.25	2.18	1.00
10 min	2.25	2.18	1.00
15 min	2.18	2.18	1.00
30 min	2.18	2.18	1.00
45 min	2.18	2.07	0.95
60 min	2.18	1.96	0.90
75 min	2.18	1.86	0.85
90 min	2.18	1.70	0.78
105 min	2.18	1.53	0.70
120 min	2.18	1.42	0.65
135 min	2.18	1.27	0.58
150 min	2.18	1.09	0.50
165 min	2.18	0.88	0.40
180 min	2.18	0.73	0.33
195 min	2.18	0.64	0.29
210 min	2.18	0.59	0.27
225 min	2.18	0.53	0.24
240 min	2.18	0.48	0.22
255 min	2.18	0.44	0.20
270 min	2.18	0.41	0.19
285 min	2.18	0.38	0.18
300 min	2.18	0.35	0.16

Table B.10 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at mixed size of chitosan

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	130.28		52.48	0.20	7.87
Initial FM 30 min	107.34		70.42	80.3	8.09
Initial FM 60 min	86.24		70.31	80.7	8.04
Initial FM 90 min	75.23		70.36	80.3	8.05
Initial FM 120 min	72.02		70.56	80.6	8.07
Initial FM 150 min	69.72		70.52	80.5	8.03
Initial FM 180 min	67.89		70.40	80.3	8.06
Initial FM 210 min	66.51		70.49	80.6	8.08
Initial FM 240 min	66.06		70.38	80.5	8.04
Initial FM 270 min	65.60		70.42	80.8	8.02
Initial FM 300 min	65.14		70.35	80.7	8.09
Permeate 30 min	102.75	21.13	70.37	80.8	8.05
Permeate 60 min	83.49	35.92	70.21	80.9	8.07
Permeate 90 min	73.39	43.66	70.42	80.1	8.03
Permeate 120 min	70.64	45.77	70.30	80.7	8.04
Permeate 150 min	68.81	47.18	70.44	80.3	8.08
Permeate 180 min	67.43	48.24	70.37	80.4	8.05
Permeate 210 min	66.06	49.30	70.42	80.8	8.03
Permeate 240 min	65.60	49.65	70.22	80.2	8.07
Permeate 270 min	65.14	50.00	70.40	80.3	8.09
Permeate 300 min	64.68	50.35	70.33	80.5	8.02

Table B.11 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at chitosan size ≥4.0 mm

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.12	1.96	1.00
5 min	2.12	1.96	1.00
10 min	2.12	1.96	1.00
15 min	2.12	1.96	1.00
30 min	2.12	1.96	1.00
45 min	2.12	1.96	1.00
60 min	2.12	1.82	0.93
75 min	2.12	1.70	0.87
90 min	2.12	1.53	0.78
105 min	1.96	1.34	0.68
120 min	1.96	1.21	0.62
135 min	1.96	1.09	0.56
150 min	1.96	0.88	0.45
165 min	1.96	0.73	0.37
180 min	1.96	0.62	0.32
195 min	1.96	0.53	0.27
210 min	1.96	0.48	0.25
225 min	1.96	0.44	0.23
240 min	1.96	0.41	0.21
255 min	1.96	0.38	0.20
270 min	1.96	0.34	0.18
285 min	1.96	0.33	0.17
300 min	1.96	0.31	0.16

Table B.12 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at chitosan size ≥4.0 mm

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.87		51.05	0.22	7.84
Initial FM 30 min	103.47		70.41	80.9	8.04
Initial FM 60 min	81.54		70.47	80.5	8.07
Initial FM 90 min	71.48		70.42	80.8	8.05
Initial FM 120 min	67.82		70.49	80.6	8.08
Initial FM 150 min	66.45		70.44	80.7	8.09
Initial FM 180 min	65.54		70.43	80.4	8.03
Initial FM 210 min	63.71		70.45	80.2	8.06
Initial FM 240 min	63.71		70.53	80.3	8.04
Initial FM 270 min	63.25		70.49	80.5	8.07
Initial FM 300 min	62.80		70.51	80.9	8.03
Permeate 30 min	98.90	21.42	70.49	80.4	8.08
Permeate 60 min	78.79	37.40	70.42	80.8	8.06
Permeate 90 min	69.65	44.66	70.47	80.2	8.04
Permeate 120 min	66.91	46.84	70.41	80.3	8.03
Permeate 150 min	66.00	47.57	70.45	80.5	8.07
Permeate 180 min	65.08	48.29	70.43	80.7	8.02
Permeate 210 min	63.25	49.75	70.50	80.6	8.08
Permeate 240 min	63.25	49.75	70.47	80.9	8.06
Permeate 270 min	62.80	50.11	70.49	80.4	8.05
Permeate 300 min	62.34	50.47	70.53	80.1	8.07

Table B.13 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at chitosan size 1.7-4.0 mm

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.18	2.12	1.00
5 min	2.18	2.12	1.00
10 min	2.18	2.12	1.00
15 min	2.18	2.12	1.00
30 min	2.18	2.12	1.00
45 min	2.18	2.12	1.00
60 min	2.18	2.12	1.00
75 min	2.18	1.91	0.90
90 min	2.18	1.70	0.80
105 min	2.18	1.53	0.72
120 min	2.18	1.34	0.63
135 min	2.18	1.21	0.57
150 min	2.18	1.09	0.51
165 min	2.18	0.88	0.41
180 min	2.12	0.73	0.34
195 min	2.12	0.64	0.30
210 min	2.12	0.59	0.28
225 min	2.12	0.53	0.25
240 min	2.12	0.46	0.22
255 min	2.12	0.41	0.19
270 min	2.12	0.38	0.18
285 min	2.12	0.34	0.16
300 min	2.12	0.31	0.15

Table B.14 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at chitosan size 1.7-4.0 mm

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.45		50.84	0.21	7.86
Initial FM 30 min	103.49		70.54	80.2	8.05
Initial FM 60 min	83.78		70.61	80.8	8.09
Initial FM 90 min	70.79		70.57	80.5	8.04
Initial FM 120 min	67.20		70.55	80.7	8.07
Initial FM 150 min	65.86		70.51	80.1	8.02
Initial FM 180 min	64.96		70.59	80.4	8.06
Initial FM 210 min	64.07		70.49	80.8	8.03
Initial FM 240 min	63.62		70.52	80.2	8.05
Initial FM 270 min	63.17		70.55	80.3	8.08
Initial FM 300 min	62.28		70.50	80.5	8.02
Permeate 30 min	98.57	21.43	70.53	80.4	8.07
Permeate 60 min	81.09	35.36	70.51	80.5	8.05
Permeate 90 min	69.00	45.00	70.48	80.1	8.09
Permeate 120 min	66.31	47.14	70.53	80.2	8.03
Permeate 150 min	65.41	47.86	70.57	80.9	8.08
Permeate 180 min	64.52	48.57	70.54	80.3	8.04
Permeate 210 min	63.62	49.29	70.61	80.7	8.05
Permeate 240 min	63.17	49.64	70.52	80.3	8.07
Permeate 270 min	62.72	50.00	70.55	80.6	8.03
Permeate 300 min	61.83	50.71	70.51	80.9	8.09

Table B.15 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at chitosan size 0.85-1.7 mm

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.18	2.07	1.00
5 min	2.18	2.07	1.00
10 min	2.18	2.07	1.00
15 min	2.18	2.07	1.00
30 min	2.18	2.07	1.00
45 min	2.18	2.07	1.00
60 min	2.07	2.07	1.00
75 min	2.07	1.86	0.90
90 min	2.07	1.66	0.80
105 min	2.07	1.42	0.69
120 min	2.07	1.21	0.59
135 min	2.07	1.02	0.49
150 min	2.07	0.82	0.40
165 min	2.07	0.64	0.31
180 min	2.07	0.59	0.28
195 min	2.07	0.53	0.26
210 min	2.07	0.50	0.24
225 min	2.07	0.48	0.23
240 min	2.07	0.42	0.21
255 min	2.07	0.41	0.20
270 min	2.07	0.34	0.17
285 min	2.07	0.31	0.15
300 min	2.07	0.27	0.13

Table B.16 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at chitosan size 0.85-1.7 mm

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	130.43		52.71	0.21	7.83
Initial FM 30 min	107.66		73.65	80.5	8.09
Initial FM 60 min	86.82		73.69	80.1	8.07
Initial FM 90 min	72.77		73.71	80.3	8.05
Initial FM 120 min	69.86		72.70	80.7	8.08
Initial FM 150 min	68.90		72.68	80.6	8.06
Initial FM 180 min	67.93		73.73	80.4	8.09
Initial FM 210 min	67.44		73.73	80.2	8.07
Initial FM 240 min	66.47		73.75	80.9	8.05
Initial FM 270 min	65.50		73.71	80.7	8.08
Initial FM 300 min	64.53		72.72	80.5	8.06
Permeate 30 min	102.33	21.55	73.69	80.8	8.05
Permeate 60 min	83.91	35.66	73.71	80.5	8.09
Permeate 90 min	70.83	45.69	73.70	80.7	8.08
Permeate 120 min	68.41	47.55	73.73	80.4	8.06
Permeate 150 min	67.93	47.92	73.75	80.3	8.09
Permeate 180 min	67.44	48.29	73.71	80.6	8.08
Permeate 210 min	66.96	48.66	73.72	80.7	8.05
Permeate 240 min	65.99	49.41	73.77	80.3	8.07
Permeate 270 min	65.02	50.15	73.73	80.5	8.06
Permeate 300 min	64.05	50.89	73.75	80.9	8.08

Table B.17 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at chitosan size ≤0.85 mm

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.12	2.07	1.00
5 min	2.12	2.07	1.00
10 min	2.12	2.07	1.00
15 min	2.12	2.07	1.00
30 min	2.12	2.07	1.00
45 min	2.12	2.07	1.00
60 min	2.12	2.07	1.00
75 min	2.07	1.86	0.90
90 min	2.07	1.68	0.81
105 min	2.07	1.44	0.70
120 min	2.07	1.23	0.60
135 min	2.07	1.02	0.49
150 min	2.07	0.82	0.40
165 min	2.07	0.64	0.31
180 min	2.07	0.53	0.26
195 min	2.07	0.50	0.24
210 min	2.07	0.48	0.23
225 min	2.07	0.42	0.21
240 min	2.07	0.41	0.20
255 min	2.07	0.34	0.17
270 min	2.07	0.31	0.15
285 min	2.07	0.27	0.13
300 min	2.07	0.23	0.11

Table B.18 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at chitosan size ≤0.85 mm

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.85		50.83	0.20	7.85
Initial FM 30 min	103.94		75.89	80.7	8.08
Initial FM 60 min	84.26		75.87	80.2	8.06
Initial FM 90 min	69.95		75.91	80.6	8.07
Initial FM 120 min	66.82		75.95	80.9	8.09
Initial FM 150 min	65.92		75.88	80.5	8.05
Initial FM 180 min	65.47		75.93	80.3	8.08
Initial FM 210 min	65.03		75.94	80.1	8.07
Initial FM 240 min	64.58		75.90	80.4	8.09
Initial FM 270 min	64.13		75.87	80.7	8.06
Initial FM 300 min	62.79		75.92	80.5	8.10
Permeate 30 min	99.02	21.32	75.91	80.3	8.09
Permeate 60 min	81.13	35.54	75.87	80.5	8.08
Permeate 90 min	68.16	45.84	75.94	80.1	8.07
Permeate 120 min	65.92	47.62	75.91	80.2	8.04
Permeate 150 min	65.47	47.97	75.87	80.8	8.06
Permeate 180 min	65.03	48.33	75.79	80.6	8.10
Permeate 210 min	64.58	48.69	75.92	80.9	8.08
Permeate 240 min	64.13	49.04	75.75	80.7	8.07
Permeate 270 min	63.69	49.40	75.83	80.2	8.09
Permeate 300 min	62.34	50.46	75.95	80.3	8.08

Table B.19 Permeate flux of formaldehyde solution by microfiltration membrane at original pH

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.18	2.18	1.00
5 min	2.18	2.18	1.00
10 min	2.18	2.18	1.00
15 min	2.18	2.18	1.00
30 min	2.18	2.18	1.00
45 min	2.18	2.18	1.00
60 min	2.18	2.18	1.00
75 min	2.18	2.18	1.00
90 min	2.18	2.18	1.00
105 min	2.18	2.18	1.00
120 min	2.18	2.18	1.00
135 min	2.18	2.18	1.00
150 min	2.18	2.18	1.00
165 min	2.18	2.18	1.00
180 min	2.18	2.07	0.95
195 min	2.18	2.07	0.95
210 min	2.18	2.07	0.95
225 min	2.18	2.07	0.95
240 min	2.18	2.07	0.95
255 min	2.18	2.07	0.95
270 min	2.18	2.07	0.95
285 min	2.18	2.07	0.95
300 min	2.18	2.07	0.95

Table B.20 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by microfiltration membrane at original pH

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.72		50.48	0.35	7.87
Initial FM 30 min	125.72		49.82	0.33	7.83
Initial FM 60 min	125.72		50.27	0.29	7.85
Initial FM 90 min	125.72		50.17	0.31	7.85
Initial FM 120 min	125.72		50.21	0.25	7.84
Initial FM 150 min	125.72		50.23	0.27	7.81
Initial FM 180 min	125.24		50.31	0.22	7.85
Initial FM 210 min	125.24		50.25	0.30	7.82
Initial FM 240 min	125.24		50.20	0.24	7.84
Initial FM 270 min	125.24		49.89	0.31	7.83
Initial FM 300 min	125.24		49.91	0.33	7.87
Permeate 30 min	125.24	0.38	49.88	0.24	7.85
Permeate 60 min	125.24	0.38	49.93	0.21	7.87
Permeate 90 min	125.24	0.38	50.73	0.35	7.89
Permeate 120 min	125.24	0.38	50.65	0.22	7.83
Permeate 150 min	125.24	0.38	49.81	0.29	7.84
Permeate 180 min	124.76	0.77	50.55	0.31	7.81
Permeate 210 min	124.76	0.77	50.63	0.34	7.82
Permeate 240 min	124.76	0.77	50.71	0.25	7.86
Permeate 270 min	124.76	0.77	49.84	0.27	7.83
Permeate 300 min	124.76	0.77	50.60	0.23	7.82

Table B.21 Permeate flux of formaldehyde solution by microfiltration membrane at pH 4

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.12	2.07	1.00
5 min	2.12	2.07	1.00
10 min	2.12	2.07	1.00
15 min	2.12	2.07	1.00
30 min	2.12	2.07	1.00
45 min	2.12	2.07	1.00
60 min	2.12	2.07	1.00
75 min	2.12	2.07	1.00
90 min	2.12	2.07	1.00
105 min	2.12	2.07	1.00
120 min	2.12	2.07	1.00
135 min	2.12	2.07	1.00
150 min	2.12	2.07	1.00
165 min	2.12	2.07	1.00
180 min	2.12	1.96	0.95
195 min	2.12	1.96	0.95
210 min	2.12	1.96	0.95
225 min	2.07	1.96	0.95
240 min	2.07	1.96	0.95
255 min	2.07	1.96	0.95
270 min	2.07	1.96	0.95
285 min	2.07	1.96	0.95
300 min	2.07	1.96	0.95

Table B.22 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by microfiltration membrane at pH 4

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	120.79		50.11	0.26	4.05
Initial FM 30 min	120.79		50.21	0.31	4.11
Initial FM 60 min	120.79		50.19	0.25	4.09
Initial FM 90 min	120.79		50.23	0.29	4.07
Initial FM 120 min	120.79		50.17	0.30	4.07
Initial FM 150 min	120.31		50.25	0.33	4.08
Initial FM 180 min	120.31		50.20	0.22	4.10
Initial FM 210 min	120.31		50.24	0.26	4.11
Initial FM 240 min	120.31		50.19	0.31	4.13
Initial FM 270 min	120.31		50.21	0.27	4.09
Initial FM 300 min	120.31		50.19	0.24	4.15
Permeate 30 min	120.31	0.40	50.27	0.21	4.11
Permeate 60 min	120.31	0.40	50.23	0.37	4.10
Permeate 90 min	120.31	0.40	50.20	0.28	4.13
Permeate 120 min	120.31	0.40	50.15	0.21	4.11
Permeate 150 min	119.83	0.79	50.17	0.33	4.10
Permeate 180 min	119.83	0.79	50.21	0.35	4.13
Permeate 210 min	119.83	0.79	50.25	0.27	4.11
Permeate 240 min	119.83	0.79	50.20	0.22	4.10
Permeate 270 min	119.83	0.79	50.22	0.29	4.11
Permeate 300 min	119.83	0.79	50.19	0.33	4.17

Table B.23 Permeate flux of formaldehyde solution by microfiltration membrane at pH 7

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.18	2.07	1.00
5 min	2.18	2.07	1.00
10 min	2.18	2.07	1.00
15 min	2.18	2.07	1.00
30 min	2.18	2.07	1.00
45 min	2.18	2.07	1.00
60 min	2.18	2.07	1.00
75 min	2.18	2.07	1.00
90 min	2.18	2.07	1.00
105 min	2.18	2.07	1.00
120 min	2.18	2.07	1.00
135 min	2.07	2.01	0.97
150 min	2.07	2.01	0.97
165 min	2.07	2.01	0.97
180 min	2.07	2.01	0.97
195 min	2.07	2.01	0.97
210 min	2.07	2.01	0.97
225 min	2.07	2.01	0.97
240 min	2.07	2.01	0.97
255 min	2.07	2.01	0.97
270 min	2.07	2.01	0.97
285 min	2.07	2.01	0.97
300 min	2.07	2.01	0.97

Table B.24 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by microfiltration membrane at pH 7

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.24		50.77	0.28	7.03
Initial FM 30 min	125.24		50.25	0.26	7.11
Initial FM 60 min	125.24		50.25	0.31	7.13
Initial FM 90 min	125.24		50.31	0.37	7.07
Initial FM 120 min	125.24		50.25	0.35	7.10
Initial FM 150 min	125.24		50.30	0.33	7.09
Initial FM 180 min	124.76		50.23	0.21	7.11
Initial FM 210 min	124.76		50.27	0.26	7.13
Initial FM 240 min	124.76		50.32	0.21	7.10
Initial FM 270 min	124.76		50.35	0.30	7.11
Initial FM 300 min	124.76		50.31	0.34	7.10
Permeate 30 min	124.76	0.38	50.27	0.27	7.09
Permeate 60 min	124.76	0.38	50.29	0.21	7.10
Permeate 90 min	124.76	0.38	50.31	0.38	7.11
Permeate 120 min	124.76	0.38	50.25	0.25	7.07
Permeate 150 min	124.76	0.38	50.27	0.34	7.13
Permeate 180 min	124.28	0.77	50.25	0.39	7.10
Permeate 210 min	124.28	0.77	50.33	0.25	7.09
Permeate 240 min	124.28	0.77	50.34	0.22	7.13
Permeate 270 min	124.28	0.77	50.31	0.31	7.10
Permeate 300 min	124.28	0.77	50.33	0.29	7.11

Table B.25 Permeate flux of formaldehyde solution by microfiltration membrane at pH 10

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.18	2.12	1.00
5 min	2.18	2.12	1.00
10 min	2.18	2.12	1.00
15 min	2.18	2.12	1.00
30 min	2.18	2.12	1.00
45 min	2.18	2.12	1.00
60 min	2.18	2.12	1.00
75 min	2.18	2.12	1.00
90 min	2.18	2.12	1.00
105 min	2.18	2.12	1.00
120 min	2.18	2.12	1.00
135 min	2.12	2.07	0.97
150 min	2.12	2.07	0.97
165 min	2.12	2.07	0.97
180 min	2.12	2.07	0.97
195 min	2.12	2.07	0.97
210 min	2.12	2.07	0.97
225 min	2.12	2.07	0.97
240 min	2.12	2.07	0.97
255 min	2.12	2.07	0.97
270 min	2.12	2.07	0.97
285 min	2.12	2.07	0.97
300 min	2.12	2.07	0.97

Table B.26 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by microfiltration membrane at pH 10

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.53		50.88	0.28	10.10
Initial FM 30 min	125.53		50.31	0.21	10.17
Initial FM 60 min	125.53		50.27	0.33	10.11
Initial FM 90 min	125.53		50.30	0.35	10.14
Initial FM 120 min	125.05		50.29	0.27	10.13
Initial FM 150 min	125.05		50.33	0.22	10.15
Initial FM 180 min	125.05		50.35	0.29	10.11
Initial FM 210 min	125.05		50.37	0.22	10.19
Initial FM 240 min	125.05		50.31	0.30	10.21
Initial FM 270 min	125.05		50.33	0.24	10.11
Initial FM 300 min	125.05		50.37	0.31	10.13
Permeate 30 min	125.05	0.38	50.34	0.33	10.13
Permeate 60 min	125.05	0.38	50.37	0.24	10.15
Permeate 90 min	125.05	0.38	50.35	0.21	10.17
Permeate 120 min	124.57	0.76	50.33	0.21	10.20
Permeate 150 min	124.57	0.76	50.31	0.30	10.21
Permeate 180 min	124.57	0.76	50.37	0.34	10.15
Permeate 210 min	124.57	0.76	50.35	0.27	10.13
Permeate 240 min	124.57	0.76	50.33	0.21	10.15
Permeate 270 min	124.57	0.76	50.34	0.33	10.11
Permeate 300 min	124.57	0.76	50.37	0.26	10.11

Table B.27 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at original pH

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.12	2.12	1.00
5 min	2.12	2.12	1.00
10 min	2.12	2.12	1.00
15 min	2.12	2.12	1.00
30 min	2.12	2.12	1.00
45 min	2.12	2.12	1.00
60 min	2.12	2.01	0.95
75 min	2.12	1.91	0.90
90 min	2.12	1.78	0.84
105 min	2.12	1.66	0.78
120 min	2.12	1.53	0.72
135 min	2.12	1.42	0.67
150 min	2.12	1.27	0.60
165 min	2.12	1.09	0.51
180 min	2.12	0.88	0.41
195 min	2.12	0.75	0.35
210 min	2.12	0.62	0.29
225 min	2.12	0.52	0.24
240 min	2.12	0.46	0.22
255 min	2.12	0.39	0.18
270 min	2.12	0.36	0.17
285 min	2.12	0.34	0.16
300 min	2.12	0.31	0.15

Table B.28 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at original pH

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	120.41		50.23	0.25	7.87
Initial FM 30 min	98.16		70.35	80.6	8.05
Initial FM 60 min	79.30		70.37	80.9	8.03
Initial FM 90 min	67.70		70.21	80.7	8.06
Initial FM 120 min	64.80		70.42	80.2	8.05
Initial FM 150 min	63.83		70.30	80.3	8.04
Initial FM 180 min	62.86		70.44	80.8	8.07
Initial FM 210 min	62.38		70.37	80.5	8.07
Initial FM 240 min	61.41		70.42	80.7	8.09
Initial FM 270 min	60.93		70.22	80.4	8.05
Initial FM 300 min	60.44		70.40	80.3	8.08
Permeate 30 min	93.81	22.09	70.33	80.6	8.06
Permeate 60 min	76.40	36.55	70.42	80.7	8.04
Permeate 90 min	66.25	44.98	70.31	80.3	8.09
Permeate 120 min	63.83	46.99	70.36	80.5	8.07
Permeate 150 min	63.35	47.39	70.56	80.9	8.05
Permeate 180 min	62.38	48.19	70.52	80.3	8.06
Permeate 210 min	61.90	48.59	70.40	80.1	8.04
Permeate 240 min	60.93	49.40	70.49	80.4	8.08
Permeate 270 min	60.44	49.80	70.38	80.7	8.04
Permeate 300 min	59.96	50.20	70.42	80.5	8.05

Table B.29 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at pH 4

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.18	2.07	1.00
5 min	2.18	2.07	1.00
10 min	2.18	2.07	1.00
15 min	2.18	2.07	1.00
30 min	2.18	2.07	1.00
45 min	2.18	2.07	1.00
60 min	2.18	1.96	0.95
75 min	2.18	1.82	0.88
90 min	2.18	1.66	0.80
105 min	2.18	1.53	0.74
120 min	2.18	1.39	0.67
135 min	2.18	1.21	0.59
150 min	2.18	1.06	0.51
165 min	2.18	0.91	0.44
180 min	2.18	0.77	0.37
195 min	2.07	0.59	0.29
210 min	2.07	0.52	0.25
225 min	2.07	0.45	0.22
240 min	2.07	0.40	0.20
255 min	2.07	0.36	0.18
270 min	2.07	0.34	0.17
285 min	2.07	0.31	0.15
300 min	2.07	0.29	0.14

Table B.30 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at pH 4

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	120.58		50.54	0.23	4.05
Initial FM 30 min	100.19		70.39	80.2	4.27
Initial FM 60 min	79.32		70.43	80.8	4.23
Initial FM 90 min	68.16		70.37	80.5	4.24
Initial FM 120 min	67.67		70.51	80.7	4.26
Initial FM 150 min	63.30		70.47	80.1	4.28
Initial FM 180 min	62.33		70.45	80.5	4.25
Initial FM 210 min	61.84		70.42	80.4	4.23
Initial FM 240 min	61.36		70.41	80.2	4.27
Initial FM 270 min	60.39		70.43	80.8	4.24
Initial FM 300 min	59.90		70.48	80.6	4.29
Permeate 30 min	95.34	20.93	70.50	80.9	4.25
Permeate 60 min	76.41	36.63	70.47	80.3	4.26
Permeate 90 min	66.21	45.09	70.48	80.1	4.28
Permeate 120 min	65.73	45.49	70.48	80.4	4.27
Permeate 150 min	62.33	48.31	70.54	80.7	4.25
Permeate 180 min	61.84	48.71	70.35	80.6	4.27
Permeate 210 min	61.36	49.11	70.51	80.7	4.26
Permeate 240 min	60.87	49.52	70.45	80.3	4.29
Permeate 270 min	59.90	50.32	70.38	80.5	4.26
Permeate 300 min	59.42	50.72	70.44	80.9	4.24

Table B.31 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at pH 7

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.18	2.12	1.00
5 min	2.18	2.12	1.00
10 min	2.18	2.12	1.00
15 min	2.18	2.12	1.00
30 min	2.18	2.12	1.00
45 min	2.18	2.12	1.00
60 min	2.18	1.99	0.94
75 min	2.18	1.89	0.89
90 min	2.18	1.68	0.79
105 min	2.18	1.50	0.71
120 min	2.18	1.32	0.62
135 min	2.18	1.21	0.57
150 min	2.18	1.02	0.48
165 min	2.12	0.82	0.39
180 min	2.12	0.73	0.34
195 min	2.12	0.69	0.32
210 min	2.12	0.57	0.27
225 min	2.12	0.49	0.23
240 min	2.12	0.42	0.20
255 min	2.12	0.36	0.17
270 min	2.12	0.34	0.16
285 min	2.12	0.31	0.15
300 min	2.12	0.29	0.14

Table B.32 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at pH 7

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	120.23		49.31	0.27	7.10
Initial FM 30 min	99.04		70.54	80.1	7.27
Initial FM 60 min	80.25		70.63	80.4	7.24
Initial FM 90 min	67.24		70.57	80.7	7.28
Initial FM 120 min	64.84		70.58	80.6	7.23
Initial FM 150 min	63.87		70.55	80.7	7.25
Initial FM 180 min	62.91		70.61	80.2	7.29
Initial FM 210 min	62.43		70.67	80.8	7.26
Initial FM 240 min	61.46		70.59	80.5	7.22
Initial FM 270 min	60.98		70.67	80.7	7.27
Initial FM 300 min	60.02		70.71	80.4	7.25
Permeate 30 min	94.22	21.63	70.65	80.8	7.29
Permeate 60 min	77.36	35.66	70.63	80.5	7.27
Permeate 90 min	65.32	45.67	70.55	80.1	7.25
Permeate 120 min	63.87	46.88	70.67	80.4	7.27
Permeate 150 min	63.39	47.28	70.61	80.3	7.28
Permeate 180 min	62.43	48.08	70.59	80.8	7.22
Permeate 210 min	61.95	48.48	70.69	80.5	7.25
Permeate 240 min	60.98	49.28	70.63	80.7	7.26
Permeate 270 min	60.50	49.68	70.57	80.3	7.23
Permeate 300 min	59.54	50.48	70.61	80.6	7.28

Table B.33 Permeate flux of formaldehyde solution by chitosan-enhanced microfiltration membrane at pH 10

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)
0 min	2.12	2.07	1.00
5 min	2.12	2.07	1.00
10 min	2.12	2.07	1.00
15 min	2.12	2.07	1.00
30 min	2.12	2.07	1.00
45 min	2.12	2.07	1.00
60 min	2.12	2.07	1.00
75 min	2.12	1.96	0.95
90 min	2.12	1.84	0.89
105 min	2.12	1.59	0.77
120 min	2.12	1.42	0.69
135 min	2.12	1.19	0.58
150 min	2.07	1.02	0.49
165 min	2.07	0.80	0.39
180 min	2.07	0.69	0.33
195 min	2.07	0.59	0.29
210 min	2.07	0.55	0.27
225 min	2.07	0.52	0.25
240 min	2.07	0.42	0.21
255 min	2.07	0.39	0.19
270 min	2.07	0.34	0.17
285 min	2.07	0.34	0.16
300 min	2.07	0.31	0.15

Table B.34 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced microfiltration membrane at pH 10

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	120.10		49.65	0.28	10.07
Initial FM 30 min	98.94		70.71	80.1	9.82
Initial FM 60 min	80.67		70.79	80.7	9.89
Initial FM 90 min	66.25		70.77	80.3	9.84
Initial FM 120 min	64.81		70.73	80.4	9.85
Initial FM 150 min	62.88		70.71	80.8	9.87
Initial FM 180 min	62.40		70.83	80.2	9.82
Initial FM 210 min	60.96		70.79	80.4	9.85
Initial FM 240 min	60.96		70.81	80.3	9.85
Initial FM 270 min	60.00		70.77	80.8	9.87
Initial FM 300 min	59.52		70.87	80.5	9.89
Permeate 30 min	94.13	21.62	70.83	80.4	9.88
Permeate 60 min	77.79	35.23	70.85	80.2	9.87
Permeate 90 min	64.81	46.04	70.79	80.6	9.88
Permeate 120 min	63.85	46.84	70.75	80.7	9.88
Permeate 150 min	62.40	48.04	70.73	80.6	9.87
Permeate 180 min	61.92	48.44	70.82	80.7	9.86
Permeate 210 min	60.48	49.64	70.78	80.2	9.82
Permeate 240 min	60.48	49.64	70.89	80.8	9.88
Permeate 270 min	59.52	50.44	70.81	80.5	9.87
Permeate 300 min	59.04	50.84	70.79	80.7	9.89

EXPERIMENT C

CHITOSAN-ENHANCED UF MEMBRANE FILTRATION

Table C.1 Permeate and retentate flux of formaldehyde solution by ultrafiltration membrane at various times

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.047	0.047	1.00	0.047
5 min	0.047	0.047	1.00	0.047
10 min	0.047	0.047	1.00	0.047
15 min	0.047	0.047	1.00	0.047
30 min	0.047	0.047	1.00	0.047
45 min	0.047	0.047	1.00	0.047
60 min	0.047	0.047	1.00	0.047
75 min	0.047	0.047	1.00	0.047
90 min	0.047	0.047	1.00	0.047
105 min	0.047	0.047	1.00	0.047
120 min	0.047	0.047	1.00	0.047
135 min	0.047	0.045	0.96	0.044
150 min	0.047	0.045	0.96	0.044
165 min	0.047	0.045	0.96	0.044
180 min	0.047	0.045	0.96	0.044
195 min	0.047	0.045	0.96	0.044
210 min	0.047	0.045	0.96	0.044
225 min	0.047	0.045	0.96	0.044
240 min	0.047	0.045	0.96	0.044
255 min	0.047	0.045	0.96	0.044
270 min	0.047	0.045	0.96	0.044
285 min	0.047	0.045	0.96	0.044
300 min	0.047	0.045	0.96	0.044

Table C.2 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by ultrafiltration membrane at various times

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.34		50.23	0.23	7.87
Initial FM 30 min	125.34		50.21	0.21	7.85
Initial FM 60 min	125.34		50.25	0.25	7.81
Initial FM 90 min	125.34		50.33	0.19	7.83
Initial FM 120 min	125.34		50.45	0.22	7.84
Initial FM 150 min	125.34		50.43	0.25	7.82
Initial FM 180 min	124.86		50.22	0.17	7.84
Initial FM 210 min	124.86		50.17	0.23	7.81
Initial FM 240 min	124.86		50.27	0.29	7.87
Initial FM 270 min	124.86		50.34	0.17	7.83
Initial FM 300 min	124.86		50.28	0.15	7.85
Permeate 30 min	124.86	0.38	50.21	0.20	7.87
Permeate 60 min	124.86	0.38	50.30	0.22	7.86
Permeate 90 min	124.38	0.77	50.56	0.24	7.85
Permeate 120 min	124.86	0.38	50.37	0.18	7.88
Permeate 150 min	124.86	0.38	50.44	0.25	7.81
Permeate 180 min	124.38	0.77	50.27	0.17	7.83
Permeate 210 min	124.38	0.77	50.44	0.23	7.83
Permeate 240 min	124.38	0.77	50.35	0.19	7.85
Permeate 270 min	124.38	0.77	50.21	0.17	7.84
Permeate 300 min	124.38	0.77	50.47	0.21	7.82

Table C.3 Formaldehyde concentration, DOC, turbidity, and pH in retentate by ultrafiltration membrane at various times

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	125.82	50.55	0.25	7.85
Retentate 60 min	125.82	50.61	0.27	7.88
Retentate 90 min	125.82	50.57	0.22	7.83
Retentate 120 min	125.82	50.55	0.19	7.87
Retentate 150 min	125.82	50.54	0.24	7.84
Retentate 180 min	124.86	50.52	0.26	7.85
Retentate 210 min	124.86	50.51	0.20	7.88
Retentate 240 min	124.86	50.50	0.15	7.87
Retentate 270 min	124.86	50.51	0.18	7.85
Retentate 300 min	124.86	50.49	0.23	7.86

Table C.4 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at chitosan:formaldehyde ratio of 100:1

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.047	0.047	1.00	0.042
5 min	0.047	0.047	1.00	0.042
10 min	0.047	0.047	1.00	0.042
15 min	0.047	0.047	1.00	0.042
30 min	0.047	0.047	1.00	0.042
45 min	0.047	0.047	1.00	0.042
60 min	0.047	0.047	1.00	0.042
75 min	0.047	0.045	0.96	0.039
90 min	0.047	0.045	0.95	0.039
105 min	0.047	0.044	0.94	0.039
120 min	0.047	0.044	0.93	0.039
135 min	0.047	0.043	0.92	0.039
150 min	0.047	0.043	0.91	0.039
165 min	0.047	0.042	0.90	0.039
180 min	0.047	0.042	0.89	0.035
195 min	0.047	0.042	0.88	0.035
210 min	0.047	0.041	0.87	0.035
225 min	0.047	0.041	0.87	0.035
240 min	0.047	0.040	0.86	0.035
255 min	0.047	0.040	0.85	0.035
270 min	0.047	0.040	0.84	0.035
285 min	0.047	0.039	0.83	0.035
300 min	0.047	0.039	0.82	0.035

Table C.5 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at chitosan:formaldehyde ratio of 100:1

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	121.50		50.14	0.23	7.84
Initial FM 30 min	95.59		50.83	0.74	7.95
Initial FM 60 min	91.75		40.87	0.57	7.98
Initial FM 90 min	86.95		40.91	0.61	8.01
Initial FM 120 min	82.63		40.95	0.89	7.96
Initial FM 150 min	81.19		40.81	0.73	7.94
Initial FM 180 min	80.23		40.93	0.68	7.98
Initial FM 210 min	79.75		40.95	0.85	7.99
Initial FM 240 min	79.27		40.89	0.77	7.95
Initial FM 270 min	78.31		40.97	0.91	7.97
Initial FM 300 min	77.83		40.88	0.83	7.95
Permeate 30 min	90.79	25.28	50.91	0.81	7.99
Permeate 60 min	87.91	27.65	40.87	0.79	7.94
Permeate 90 min	84.07	30.81	40.91	0.58	7.97
Permeate 120 min	80.71	33.57	40.89	0.63	7.95
Permeate 150 min	80.23	33.97	40.79	0.96	7.93
Permeate 180 min	79.75	34.36	40.87	0.82	7.98
Permeate 210 min	79.27	34.76	40.83	0.97	7.97
Permeate 240 min	78.79	35.15	40.91	0.89	7.95
Permeate 270 min	77.83	35.94	40.94	0.68	7.94
Permeate 300 min	77.35	36.33	40.87	0.56	7.96

Table C.6 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at chitosan:formaldehyde ratio of 100:1

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	100.38	40.72	11.7	7.67
Retentate 60 min	90.79	30.63	10.6	7.68
Retentate 90 min	90.31	30.87	11.5	7.67
Retentate 120 min	90.79	30.95	11.3	7.66
Retentate 150 min	80.23	30.77	10.8	7.67
Retentate 180 min	80.71	30.54	10.9	7.68
Retentate 210 min	80.71	30.69	10.7	7.66
Retentate 240 min	80.23	30.88	11.9	7.67
Retentate 270 min	80.71	30.71	11.6	7.68
Retentate 300 min	80.23	30.87	11.4	7.67

Table C.7 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at chitosan:formaldehyde ratio of 200:1

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.047	0.046	1.00	0.042
5 min	0.047	0.046	1.00	0.042
10 min	0.047	0.046	1.00	0.042
15 min	0.047	0.046	1.00	0.042
30 min	0.047	0.043	0.94	0.042
45 min	0.047	0.040	0.87	0.035
60 min	0.047	0.037	0.79	0.035
75 min	0.047	0.034	0.73	0.028
90 min	0.047	0.031	0.67	0.028
105 min	0.046	0.027	0.59	0.021
120 min	0.046	0.023	0.50	0.021
135 min	0.046	0.021	0.45	0.018
150 min	0.046	0.019	0.40	0.018
165 min	0.046	0.016	0.34	0.011
180 min	0.046	0.014	0.29	0.007
195 min	0.046	0.011	0.24	0.007
210 min	0.046	0.009	0.20	0.007
225 min	0.046	0.008	0.17	0.004
240 min	0.046	0.006	0.14	0.004
255 min	0.046	0.006	0.13	0.004
270 min	0.046	0.006	0.12	0.004
285 min	0.046	0.005	0.11	0.004
300 min	0.046	0.005	0.10	0.004

Table C.8 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at chitosan:formaldehyde ratio of 200:1

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.22		51.19	0.23	7.87
Initial FM 30 min	104.20		60.78	0.87	8.03
Initial FM 60 min	84.06		50.65	0.95	8.05
Initial FM 90 min	70.49		50.81	0.73	8.07
Initial FM 120 min	67.43		50.99	0.48	8.04
Initial FM 150 min	65.67		50.72	0.61	8.08
Initial FM 180 min	64.36		50.84	0.59	8.05
Initial FM 210 min	63.05		50.84	0.78	8.07
Initial FM 240 min	62.17		50.79	0.97	8.06
Initial FM 270 min	61.73		50.91	0.55	8.08
Initial FM 300 min	61.73		50.73	0.72	8.07
Permeate 30 min	98.95	20.98	60.83	0.68	8.06
Permeate 60 min	80.56	35.66	50.75	0.54	8.09
Permeate 90 min	68.30	45.45	50.87	0.62	8.05
Permeate 120 min	65.67	47.55	50.79	0.77	8.07
Permeate 150 min	64.36	48.60	50.93	0.87	8.05
Permeate 180 min	63.49	49.30	50.88	0.74	8.04
Permeate 210 min	62.61	50.00	50.54	0.63	8.07
Permeate 240 min	61.73	50.70	50.69	0.72	8.06
Permeate 270 min	61.30	51.05	50.72	0.98	8.07
Permeate 300 min	61.30	51.05	50.61	0.85	8.08

Table C.9 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at chitosan:formaldehyde ratio of 200:1

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	100.26	50.81	19.9	7.47
Retentate 60 min	90.63	40.54	20.3	7.48
Retentate 90 min	80.56	40.63	20.7	7.48
Retentate 120 min	80.12	40.78	21.5	7.47
Retentate 150 min	71.37	40.85	21.4	7.45
Retentate 180 min	70.93	40.53	20.8	7.47
Retentate 210 min	70.93	40.84	21.7	7.48
Retentate 240 min	70.49	40.67	19.3	7.48
Retentate 270 min	70.49	40.76	21.5	7.47
Retentate 300 min	70.49	40.93	20.6	7.47

Table C.10 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at chitosan:formaldehyde ratio of 300:1

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.046	0.045	1.00	1.00
5 min	0.046	0.039	0.85	0.85
10 min	0.046	0.028	0.63	0.63
15 min	0.046	0.014	0.31	0.63
30 min	0.046	0.009	0.20	0.63
45 min	0.046	0.007	0.15	0.39
60 min	0.046	0.005	0.10	0.39
75 min	0.046	0.004	0.08	0.24
90 min	0.046	0.002	0.05	0.15
105 min	0.046	0.001	0.03	0.15
120 min	0.046	0.001	0.01	0.08

Table C.11 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at chitosan:formaldehyde ratio of 300:1

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.04		51.27	0.19	7.85
Initial FM 30 min	84.56		70.87	0.33	8.18
Initial FM 60 min	52.70		60.73	0.68	8.15
Initial FM 90 min	51.57		60.59	0.56	8.17
Initial FM 120 min	50.82		60.83	0.37	8.16
Permeate 30 min	80.43	35.67	70.75	0.74	8.15
Permeate 60 min	50.82	59.35	60.84	0.49	8.17
Permeate 90 min	50.45	59.65	60.81	0.81	8.19
Permeate 120 min	50.07	59.95	60.63	0.92	8.17

Table C.12 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at chitosan:formaldehyde ratio of 300:1

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	100.67	60.78	33.9	7.25
Retentate 60 min	90.55	50.84	31.3	7.27
Retentate 90 min	70.69	50.52	33.6	7.25
Retentate 120 min	60.94	50.95	30.4	7.26

Table C.13 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at mixed size of chitosan

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.047	0.046	1.00	0.042
5 min	0.047	0.046	1.00	0.042
10 min	0.047	0.046	1.00	0.042
15 min	0.047	0.046	1.00	0.042
30 min	0.047	0.044	0.95	0.042
45 min	0.047	0.040	0.87	0.035
60 min	0.047	0.037	0.80	0.035
75 min	0.047	0.034	0.73	0.025
90 min	0.047	0.031	0.67	0.025
105 min	0.047	0.027	0.59	0.021
120 min	0.047	0.023	0.50	0.021
135 min	0.046	0.021	0.45	0.014
150 min	0.046	0.019	0.40	0.014
165 min	0.046	0.016	0.35	0.014
180 min	0.046	0.014	0.30	0.007
195 min	0.046	0.012	0.25	0.007
210 min	0.046	0.009	0.20	0.004
225 min	0.046	0.008	0.17	0.004
240 min	0.046	0.007	0.15	0.004
255 min	0.046	0.006	0.14	0.004
270 min	0.046	0.006	0.13	0.004
285 min	0.046	0.006	0.12	0.004
300 min	0.046	0.005	0.11	0.004

Table C.14 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at mixed size of chitosan

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.59		50.55	0.18	7.87
Initial FM 30 min	104.11		60.67	0.81	8.07
Initial FM 60 min	84.00		50.82	0.92	8.02
Initial FM 90 min	70.75		50.81	0.56	8.05
Initial FM 120 min	68.92		50.99	0.37	8.04
Initial FM 150 min	66.18		50.76	0.74	8.09
Initial FM 180 min	65.72		50.94	0.63	8.05
Initial FM 210 min	63.89		50.62	0.72	8.03
Initial FM 240 min	63.89		50.75	0.98	8.05
Initial FM 270 min	62.98		50.91	0.85	8.02
Initial FM 300 min	62.07		50.84	0.49	8.05
Permeate 30 min	99.09	21.11	60.83	0.77	8.06
Permeate 60 min	81.26	35.30	50.97	0.51	8.06
Permeate 90 min	68.92	45.12	50.72	0.82	8.08
Permeate 120 min	68.01	45.85	50.57	0.68	8.04
Permeate 150 min	65.72	47.67	50.89	0.43	8.07
Permeate 180 min	65.27	48.03	50.62	0.95	8.05
Permeate 210 min	63.44	49.49	50.84	0.39	8.04
Permeate 240 min	63.44	49.49	50.78	0.63	8.03
Permeate 270 min	62.52	50.22	50.82	0.44	8.09
Permeate 300 min	61.61	50.95	50.67	0.57	8.07

Table C.15 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at mixed size of chitosan

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	101.37	60.53	21.9	8.47
Retentate 60 min	90.86	60.74	22.3	8.45
Retentate 90 min	80.80	60.83	22.4	8.45
Retentate 120 min	80.35	60.95	22.2	8.45
Retentate 150 min	71.66	60.76	22.7	8.47
Retentate 180 min	71.66	60.68	21.9	8.45
Retentate 210 min	71.21	60.82	22.0	8.45
Retentate 240 min	71.21	60.58	21.6	8.45
Retentate 270 min	70.75	60.72	21.5	8.47
Retentate 300 min	70.75	60.88	21.8	8.47

Table C.16 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at chitosan size ≥4.0 mm

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.046	0.045	1.00	1.00
5 min	0.046	0.045	1.00	1.00
10 min	0.046	0.045	1.00	1.00
15 min	0.046	0.044	0.97	1.00
30 min	0.046	0.042	0.94	1.00
45 min	0.046	0.039	0.87	0.73
60 min	0.046	0.036	0.80	0.73
75 min	0.046	0.034	0.75	0.73
90 min	0.046	0.031	0.69	0.55
105 min	0.046	0.028	0.62	0.55
120 min	0.046	0.024	0.53	0.55
135 min	0.046	0.021	0.47	0.36
150 min	0.046	0.019	0.41	0.36
165 min	0.046	0.016	0.36	0.28
180 min	0.046	0.014	0.31	0.28
195 min	0.046	0.012	0.27	0.18
210 min	0.045	0.010	0.23	0.18
225 min	0.045	0.009	0.20	0.09
240 min	0.045	0.008	0.18	0.09
255 min	0.045	0.007	0.16	0.09
270 min	0.045	0.007	0.15	0.09
285 min	0.045	0.006	0.14	0.09
300 min	0.045	0.006	0.13	0.09

Table C.17 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at chitosan size ≥4.0 mm

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	123.33		50.82	0.20	7.89
Initial FM 30 min	102.38		60.56	0.31	8.06
Initial FM 60 min	82.38		50.85	0.57	8.08
Initial FM 90 min	68.10		50.73	0.84	8.03
Initial FM 120 min	67.14		50.96	0.69	8.09
Initial FM 150 min	66.19		50.69	0.93	8.05
Initial FM 180 min	64.29		50.55	0.44	8.04
Initial FM 210 min	63.81		50.79	0.78	8.08
Initial FM 240 min	63.33		50.62	0.55	8.07
Initial FM 270 min	61.90		50.87	0.39	8.06
Initial FM 300 min	61.43		50.77	0.64	8.05
Permeate 30 min	97.62	20.85	60.89	0.85	8.09
Permeate 60 min	79.52	35.52	50.78	0.49	8.04
Permeate 90 min	66.67	45.95	50.67	0.29	8.07
Permeate 120 min	66.19	46.33	50.92	0.63	8.03
Permeate 150 min	65.71	46.72	50.59	0.96	8.06
Permeate 180 min	63.81	48.26	50.73	0.82	8.05
Permeate 210 min	63.33	48.65	50.65	0.97	8.07
Permeate 240 min	62.86	49.03	50.54	0.89	8.09
Permeate 270 min	61.43	50.19	50.82	0.68	8.04
Permeate 300 min	60.95	50.58	50.48	0.56	8.06

Table C.18 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at chitosan size ≥4.0 mm

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	100.48	50.83	20.6	8.45
Retentate 60 min	90.95	40.71	21.1	8.49
Retentate 90 min	80.48	40.64	21.5	8.47
Retentate 120 min	80.00	40.85	20.4	8.45
Retentate 150 min	70.95	40.73	20.9	8.49
Retentate 180 min	70.48	40.98	21.3	8.48
Retentate 210 min	70.95	40.58	20.7	8.45
Retentate 240 min	70.48	40.68	20.2	8.46
Retentate 270 min	70.00	40.83	20.5	8.49
Retentate 300 min	69.52	40.89	21.8	8.49

Table C.19 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at chitosan size 1.7-4.0 mm

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.046	0.046	1.00	1.00
5 min	0.046	0.046	1.00	1.00
10 min	0.046	0.046	1.00	1.00
15 min	0.046	0.046	1.00	1.00
30 min	0.046	0.043	0.94	1.00
45 min	0.046	0.041	0.89	0.92
60 min	0.046	0.039	0.84	0.92
75 min	0.046	0.037	0.79	0.82
90 min	0.046	0.034	0.73	0.65
105 min	0.046	0.031	0.67	0.65
120 min	0.046	0.026	0.58	0.65
135 min	0.046	0.023	0.49	0.28
150 min	0.046	0.019	0.41	0.28
165 min	0.046	0.015	0.32	0.28
180 min	0.046	0.013	0.28	0.28
195 min	0.046	0.012	0.26	0.09
210 min	0.046	0.011	0.23	0.09
225 min	0.046	0.010	0.21	0.09
240 min	0.046	0.009	0.19	0.09
255 min	0.046	0.008	0.16	0.09
270 min	0.046	0.006	0.13	0.09
285 min	0.046	0.006	0.12	0.09
300 min	0.046	0.005	0.11	0.09

Table C.20 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at chitosan size 1.7-4.0 mm

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	120.72		50.27	0.21	7.85
Initial FM 30 min	100.94		60.48	0.95	8.04
Initial FM 60 min	80.70		50.55	0.39	8.05
Initial FM 90 min	66.57		50.62	0.63	8.03
Initial FM 120 min	64.22		50.84	0.44	8.06
Initial FM 150 min	63.28		50.73	0.57	8.05
Initial FM 180 min	62.34		50.97	0.82	8.07
Initial FM 210 min	61.39		50.54	0.97	8.08
Initial FM 240 min	60.92		50.68	0.89	8.05
Initial FM 270 min	60.45		50.74	0.68	8.07
Initial FM 300 min	59.98		50.83	0.56	8.03
Permeate 30 min	96.23	20.28	60.91	0.44	8.04
Permeate 60 min	77.87	35.49	50.83	0.78	8.03
Permeate 90 min	64.69	46.41	50.67	0.55	8.02
Permeate 120 min	63.28	47.58	50.74	0.39	8.07
Permeate 150 min	62.81	47.97	50.98	0.64	8.05
Permeate 180 min	61.86	48.75	50.77	0.68	8.03
Permeate 210 min	60.92	49.53	50.67	0.85	8.06
Permeate 240 min	60.45	49.92	50.82	0.77	8.04
Permeate 270 min	59.98	50.31	50.79	0.91	8.07
Permeate 300 min	59.51	50.70	50.85	0.83	8.09

Table C.21 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at chitosan size 1.7-4.0 mm

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	104.71	50.83	20.1	8.45
Retentate 60 min	91.53	40.71	20.8	8.49
Retentate 90 min	80.70	40.64	21.6	8.47
Retentate 120 min	80.23	40.85	21.4	8.45
Retentate 150 min	69.87	40.73	21.5	8.49
Retentate 180 min	69.87	40.98	20.8	8.48
Retentate 210 min	69.87	40.58	20.3	8.45
Retentate 240 min	69.40	40.68	21.7	8.46
Retentate 270 min	69.87	40.83	21.2	8.49
Retentate 300 min	69.40	40.89	21.9	8.49

Table C.22 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at chitosan size 0.85-1.7 mm

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.047	0.045	1.00	1.00
5 min	0.047	0.045	1.00	1.00
10 min	0.047	0.045	1.00	1.00
15 min	0.047	0.045	1.00	1.00
30 min	0.047	0.042	0.94	1.00
45 min	0.047	0.040	0.89	0.83
60 min	0.047	0.037	0.83	0.83
75 min	0.047	0.035	0.77	0.83
90 min	0.047	0.032	0.71	0.64
105 min	0.047	0.029	0.65	0.64
120 min	0.047	0.026	0.59	0.64
135 min	0.045	0.024	0.53	0.46
150 min	0.045	0.021	0.47	0.46
165 min	0.045	0.019	0.41	0.46
180 min	0.045	0.016	0.35	0.28
195 min	0.045	0.013	0.29	0.18
210 min	0.045	0.011	0.24	0.18
225 min	0.045	0.009	0.19	0.09
240 min	0.045	0.007	0.16	0.09
255 min	0.045	0.006	0.14	0.09
270 min	0.045	0.005	0.12	0.09
285 min	0.045	0.005	0.11	0.09
300 min	0.045	0.005	0.10	0.09

Table C.23 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at chitosan size 0.85-1.7 mm

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.28		50.71	0.22	7.85
Initial FM 30 min	103.98		60.96	0.63	8.06
Initial FM 60 min	84.07		50.63	0.96	8.05
Initial FM 90 min	70.65		50.87	0.82	8.07
Initial FM 120 min	68.80		50.74	0.97	8.08
Initial FM 150 min	66.48		50.55	0.89	8.05
Initial FM 180 min	66.02		50.81	0.68	8.07
Initial FM 210 min	64.17		50.73	0.56	8.05
Initial FM 240 min	63.70		50.95	0.85	8.04
Initial FM 270 min	62.78		50.77	0.73	8.05
Initial FM 300 min	62.31		50.58	0.97	8.06
Permeate 30 min	98.89	21.06	60.85	0.89	8.07
Permeate 60 min	80.83	35.48	50.73	0.78	8.05
Permeate 90 min	68.80	45.08	50.97	0.67	8.03
Permeate 120 min	67.87	45.82	50.56	0.92	8.06
Permeate 150 min	66.02	47.30	50.64	0.59	8.04
Permeate 180 min	65.56	47.67	50.98	0.73	8.07
Permeate 210 min	63.70	49.15	50.72	0.65	8.05
Permeate 240 min	63.24	49.52	50.59	0.54	8.06
Permeate 270 min	62.31	50.26	50.94	0.82	8.09
Permeate 300 min	61.85	50.63	50.78	0.48	8.03

Table C.24 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at chitosan size 0.85-1.7 mm

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	107.69	50.79	23.9	7.89
Retentate 60 min	93.33	40.93	22.7	7.87
Retentate 90 min	81.30	40.85	23.3	7.85
Retentate 120 min	80.83	40.77	23.5	7.87
Retentate 150 min	70.65	40.53	22.8	7.89
Retentate 180 min	70.65	40.72	22.3	7.88
Retentate 210 min	70.65	40.65	23.4	7.85
Retentate 240 min	70.65	40.84	23.6	7.89
Retentate 270 min	70.65	40.51	22.8	7.88
Retentate 300 min	70.19	40.87	23.1	7.88

Table C.25 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at chitosan size ≤0.85 mm

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.046	0.045	1.00	1.00
5 min	0.046	0.045	1.00	1.00
10 min	0.046	0.045	1.00	1.00
15 min	0.046	0.045	1.00	1.00
30 min	0.046	0.043	0.95	1.00
45 min	0.046	0.041	0.90	0.83
60 min	0.046	0.039	0.85	0.83
75 min	0.046	0.036	0.79	0.65
90 min	0.046	0.033	0.73	0.65
105 min	0.046	0.030	0.66	0.65
120 min	0.046	0.027	0.60	0.46
135 min	0.046	0.024	0.54	0.46
150 min	0.046	0.021	0.47	0.37
165 min	0.045	0.019	0.41	0.37
180 min	0.045	0.016	0.35	0.37
195 min	0.045	0.013	0.29	0.18
210 min	0.045	0.012	0.26	0.18
225 min	0.045	0.010	0.23	0.09
240 min	0.045	0.009	0.20	0.09
255 min	0.045	0.007	0.16	0.09
270 min	0.045	0.006	0.14	0.09
285 min	0.045	0.005	0.12	0.09
300 min	0.045	0.005	0.10	0.09

Table C.26 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at chitosan size ≤0.85 mm

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	122.56		50.74	0.23	7.85
Initial FM 30 min	101.84		60.43	0.75	8.03
Initial FM 60 min	82.04		50.81	0.84	8.05
Initial FM 90 min	69.15		50.99	0.81	8.03
Initial FM 120 min	67.77		50.76	0.63	8.04
Initial FM 150 min	65.01		50.94	0.73	8.07
Initial FM 180 min	64.55		50.62	0.68	8.08
Initial FM 210 min	62.71		50.75	0.85	8.07
Initial FM 240 min	61.79		50.59	0.77	8.06
Initial FM 270 min	61.33		50.73	0.91	8.05
Initial FM 300 min	60.87		50.65	0.83	8.05
Permeate 30 min	96.78	21.04	60.83	0.81	8.08
Permeate 60 min	78.82	35.69	50.73	0.92	8.07
Permeate 90 min	67.31	45.08	50.95	0.56	8.06
Permeate 120 min	66.85	45.45	60.77	0.37	8.05
Permeate 150 min	64.55	47.33	50.55	0.94	8.07
Permeate 180 min	64.09	47.71	50.62	0.83	8.08
Permeate 210 min	62.25	49.21	50.84	0.97	8.09
Permeate 240 min	61.33	49.96	50.73	0.72	8.08
Permeate 270 min	60.87	50.34	50.97	0.57	8.07
Permeate 300 min	60.41	50.71	50.54	0.89	8.06

Table C.27 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at chitosan size ≤0.85 mm

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	106.91	50.68	25.3	7.47
Retentate 60 min	90.33	40.81	25.7	7.47
Retentate 90 min	81.12	40.94	23.4	7.48
Retentate 120 min	80.20	40.58	23.9	7.48
Retentate 150 min	72.84	40.77	25.1	7.45
Retentate 180 min	71.92	40.62	25.4	7.47
Retentate 210 min	71.45	40.59	25.7	7.45
Retentate 240 min	70.53	40.72	23.8	7.46
Retentate 270 min	70.07	40.91	25.5	7.47
Retentate 300 min	70.99	40.80	25.2	7.45

Table C.28 Permeate and retentate flux of formaldehyde solution by ultrafiltration membrane at original pH

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.047	0.047	1.00	0.046
5 min	0.047	0.047	1.00	0.046
10 min	0.047	0.047	1.00	0.046
15 min	0.047	0.047	1.00	0.046
30 min	0.047	0.047	1.00	0.045
45 min	0.047	0.047	1.00	0.045
60 min	0.047	0.047	1.00	0.045
75 min	0.047	0.047	1.00	0.045
90 min	0.047	0.047	1.00	0.045
105 min	0.047	0.047	1.00	0.045
120 min	0.047	0.046	0.98	0.045
135 min	0.047	0.046	0.98	0.045
150 min	0.047	0.046	0.98	0.045
165 min	0.047	0.046	0.98	0.045
180 min	0.047	0.046	0.98	0.045
195 min	0.047	0.046	0.98	0.045
210 min	0.047	0.046	0.98	0.045
225 min	0.047	0.046	0.98	0.045
240 min	0.047	0.046	0.98	0.045
255 min	0.047	0.046	0.98	0.045
270 min	0.047	0.046	0.98	0.045
285 min	0.047	0.046	0.98	0.045
300 min	0.047	0.046	0.98	0.045

Table C.29 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by ultrafiltration membrane at original pH

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.77		50.82	0.23	7.84
Initial FM 30 min	125.77		50.71	0.31	7.85
Initial FM 60 min	125.77		50.54	0.29	7.85
Initial FM 90 min	125.77		50.83	0.25	7.83
Initial FM 120 min	125.77		50.90	0.30	7.86
Initial FM 150 min	125.77		50.64	0.21	7.87
Initial FM 180 min	125.77		50.92	0.28	7.84
Initial FM 210 min	125.29		50.71	0.24	7.85
Initial FM 240 min	125.29		50.52	0.30	7.86
Initial FM 270 min	125.29		50.74	0.23	7.87
Initial FM 300 min	125.29		50.86	0.18	7.85
Permeate 30 min	125.29	0.38	50.81	0.24	7.85
Permeate 60 min	125.29	0.38	50.93	0.18	7.82
Permeate 90 min	125.29	0.38	50.65	0.26	7.84
Permeate 120 min	125.29	0.38	50.73	0.15	7.81
Permeate 150 min	125.29	0.38	50.84	0.17	7.89
Permeate 180 min	125.29	0.38	50.72	0.22	7.83
Permeate 210 min	124.81	0.76	50.79	0.13	7.83
Permeate 240 min	124.81	0.76	50.89	0.27	7.86
Permeate 270 min	124.81	0.76	50.93	0.19	7.85
Permeate 300 min	124.81	0.76	50.75	0.25	7.87

Table C.30 Formaldehyde concentration, DOC, turbidity, and pH in retentate by ultrafiltration membrane at original pH

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	125.77	55.82	0.25	7.88
Retentate 60 min	125.77	55.95	0.24	7.89
Retentate 90 min	125.77	55.87	0.25	7.87
Retentate 120 min	125.77	55.91	0.27	7.87
Retentate 150 min	125.77	55.95	0.26	7.89
Retentate 180 min	125.77	55.97	0.31	7.88
Retentate 210 min	125.77	55.77	0.33	7.88
Retentate 240 min	125.77	55.96	0.27	7.87
Retentate 270 min	125.77	55.89	0.23	7.89
Retentate 300 min	125.77	55.54	0.26	7.88

Table C.31 Permeate and retentate flux of formaldehyde solution by ultrafiltration membrane at pH 4

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.047	0.047	1.00	1.00
5 min	0.047	0.047	1.00	1.00
10 min	0.047	0.047	1.00	1.00
15 min	0.047	0.047	1.00	1.00
30 min	0.047	0.047	1.00	1.00
45 min	0.047	0.047	1.00	0.98
60 min	0.047	0.047	1.00	0.98
75 min	0.047	0.047	1.00	0.98
90 min	0.047	0.045	0.96	0.98
105 min	0.047	0.045	0.96	0.98
120 min	0.047	0.045	0.96	0.98
135 min	0.047	0.045	0.96	0.98
150 min	0.047	0.045	0.96	0.98
165 min	0.047	0.045	0.96	0.98
180 min	0.047	0.045	0.96	0.98
195 min	0.047	0.045	0.96	0.98
210 min	0.047	0.045	0.96	0.98
225 min	0.047	0.045	0.96	0.98
240 min	0.047	0.045	0.96	0.98
255 min	0.047	0.045	0.96	0.98
270 min	0.047	0.045	0.96	0.98
285 min	0.047	0.045	0.96	0.98
300 min	0.047	0.045	0.96	0.98

Table C.32 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by ultrafiltration membrane at pH 4

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	120.97		49.11	0.26	4.07
Initial FM 30 min	120.97		49.21	0.29	4.15
Initial FM 60 min	120.97		49.25	0.24	4.13
Initial FM 90 min	120.97		49.23	0.18	4.17
Initial FM 120 min	120.97		49.20	0.26	4.15
Initial FM 150 min	120.97		49.23	0.25	4.15
Initial FM 180 min	120.49		49.27	0.25	4.18
Initial FM 210 min	120.49		49.30	0.27	4.17
Initial FM 240 min	120.49		49.33	0.28	4.13
Initial FM 270 min	120.49		49.21	0.34	4.13
Initial FM 300 min	120.49		49.22	0.35	4.15
Permeate 30 min	120.49	0.39	49.31	0.37	4.15
Permeate 60 min	120.49	0.39	49.34	0.29	4.14
Permeate 90 min	120.49	0.39	49.33	0.25	4.17
Permeate 120 min	120.49	0.39	49.41	0.30	4.13
Permeate 150 min	120.49	0.39	49.39	0.29	4.17
Permeate 180 min	120.02	0.78	49.5	0.25	4.13
Permeate 210 min	120.02	0.78	49.38	0.26	4.14
Permeate 240 min	120.02	0.78	49.27	0.17	4.16
Permeate 270 min	120.02	0.78	49.33	0.19	4.15
Permeate 300 min	120.02	0.78	49.44	0.18	4.14

Table C.33 Formaldehyde concentration, DOC, turbidity, and pH in retentate by ultrafiltration membrane at pH 4

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	120.97	49.49	0.27	4.15
Retentate 60 min	120.97	49.37	0.33	4.17
Retentate 90 min	120.97	49.25	0.24	4.18
Retentate 120 min	120.97	49.44	0.22	4.17
Retentate 150 min	120.97	49.35	0.25	4.15
Retentate 180 min	120.97	49.38	0.29	4.19
Retentate 210 min	120.97	49.41	0.21	4.19
Retentate 240 min	120.97	49.47	0.26	4.2
Retentate 270 min	120.97	49.33	0.22	4.18
Retentate 300 min	120.97	49.45	0.29	4.17

Table C.34 Permeate and retentate flux of formaldehyde solution by ultrafiltration membrane at pH 7

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.046	0.046	1.00	0.045
5 min	0.046	0.046	1.00	0.045
10 min	0.046	0.046	1.00	0.045
15 min	0.046	0.046	1.00	0.045
30 min	0.046	0.046	1.00	0.045
45 min	0.046	0.046	1.00	0.044
60 min	0.046	0.046	1.00	0.044
75 min	0.046	0.046	1.00	0.044
90 min	0.046	0.046	1.00	0.044
105 min	0.046	0.046	1.00	0.044
120 min	0.046	0.046	1.00	0.044
135 min	0.046	0.046	1.00	0.044
150 min	0.046	0.045	0.98	0.044
165 min	0.046	0.045	0.98	0.044
180 min	0.046	0.045	0.98	0.044
195 min	0.046	0.045	0.98	0.044
210 min	0.046	0.045	0.98	0.044
225 min	0.046	0.045	0.98	0.044
240 min	0.046	0.045	0.98	0.044
255 min	0.046	0.045	0.98	0.044
270 min	0.046	0.045	0.98	0.044
285 min	0.046	0.045	0.98	0.044
300 min	0.046	0.045	0.98	0.044

Table C.35 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by ultrafiltration membrane at pH 7

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.56		50.75	0.27	7.07
Initial FM 30 min	125.56		50.33	0.25	7.12
Initial FM 60 min	125.56		50.32	0.31	7.14
Initial FM 90 min	125.56		50.34	0.29	7.11
Initial FM 120 min	125.56		50.31	0.23	7.11
Initial FM 150 min	125.56		50.35	0.18	7.14
Initial FM 180 min	125.09		50.32	0.21	7.15
Initial FM 210 min	125.09		50.35	0.30	7.13
Initial FM 240 min	125.09		50.34	0.29	7.14
Initial FM 270 min	125.09		50.32	0.22	7.15
Initial FM 300 min	125.09		50.33	0.25	7.12
Permeate 30 min	125.09	0.37	50.34	0.30	7.11
Permeate 60 min	125.09	0.37	50.35	0.21	7.15
Permeate 90 min	125.09	0.37	50.34	0.27	7.13
Permeate 120 min	125.09	0.37	50.37	0.24	7.11
Permeate 150 min	125.09	0.37	50.32	0.13	7.12
Permeate 180 min	124.63	0.74	50.36	0.22	7.13
Permeate 210 min	124.63	0.74	50.35	0.31	7.11
Permeate 240 min	124.63	0.74	50.35	0.25	7.13
Permeate 270 min	124.63	0.74	50.36	0.23	7.14
Permeate 300 min	124.63	0.74	50.36	0.29	7.12

Table C.36 Formaldehyde concentration, DOC, turbidity, and pH in retentate by ultrafiltration membrane at pH 7

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	125.56	50.45	0.22	7.12
Retentate 60 min	125.56	50.41	0.31	7.14
Retentate 90 min	125.56	50.43	0.25	7.12
Retentate 120 min	125.09	50.49	0.27	7.12
Retentate 150 min	125.56	50.46	0.30	7.13
Retentate 180 min	125.56	50.45	0.27	7.14
Retentate 210 min	125.09	50.46	0.25	7.12
Retentate 240 min	125.56	50.44	0.28	7.15
Retentate 270 min	125.09	50.48	0.24	7.13
Retentate 300 min	125.09	50.43	0.31	7.15

Table C.37 Permeate and retentate flux of formaldehyde solution by ultrafiltration membrane at pH 10

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.047	0.046	1.00	0.045
5 min	0.047	0.046	1.00	0.045
10 min	0.047	0.046	1.00	0.045
15 min	0.047	0.046	1.00	0.045
30 min	0.047	0.046	1.00	0.045
45 min	0.047	0.046	1.00	0.044
60 min	0.047	0.046	1.00	0.044
75 min	0.047	0.046	1.00	0.044
90 min	0.047	0.046	1.00	0.044
105 min	0.047	0.046	1.00	0.044
120 min	0.047	0.046	1.00	0.044
135 min	0.047	0.045	1.00	0.044
150 min	0.047	0.045	0.98	0.044
165 min	0.047	0.045	0.98	0.044
180 min	0.046	0.045	0.98	0.044
195 min	0.046	0.045	0.98	0.044
210 min	0.046	0.045	0.98	0.044
225 min	0.046	0.045	0.98	0.044
240 min	0.046	0.045	0.98	0.044
255 min	0.046	0.045	0.98	0.044
270 min	0.046	0.045	0.98	0.044
285 min	0.046	0.045	0.98	0.044
300 min	0.046	0.045	0.98	0.044

Table C.38 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by ultrafiltration membrane at pH 10

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	120.22		50.24	0.23	10.07
Initial FM 30 min	120.22		50.34	0.27	10.15
Initial FM 60 min	120.22		50.35	0.25	10.14
Initial FM 90 min	119.77		50.34	0.30	10.13
Initial FM 120 min	119.77		50.37	0.28	10.12
Initial FM 150 min	119.77		50.34	0.27	10.15
Initial FM 180 min	119.77		50.36	0.31	10.12
Initial FM 210 min	119.77		50.35	0.23	10.13
Initial FM 240 min	119.77		50.35	0.25	10.14
Initial FM 270 min	119.77		50.36	0.32	10.12
Initial FM 300 min	119.77		50.36	0.34	10.13
Permeate 30 min	119.77	0.38	50.33	0.29	10.13
Permeate 60 min	119.77	0.38	50.38	0.20	10.15
Permeate 90 min	119.31	0.75	50.36	0.24	10.13
Permeate 120 min	119.31	0.75	50.37	0.28	10.14
Permeate 150 min	119.31	0.75	50.35	0.21	10.12
Permeate 180 min	119.31	0.75	50.35	0.30	10.13
Permeate 210 min	119.31	0.75	50.35	0.34	10.13
Permeate 240 min	119.31	0.75	50.38	0.25	10.15
Permeate 270 min	119.31	0.75	50.34	0.25	10.14
Permeate 300 min	119.31	0.75	50.37	0.22	10.18

Table C.39 Formaldehyde concentration, DOC, turbidity, and pH in retentate at pH 10 of ultrafiltration membrane

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	119.31	50.49	0.19	10.14
Retentate 60 min	119.77	50.44	0.23	10.15
Retentate 90 min	119.31	50.47	0.30	10.15
Retentate 120 min	119.77	50.48	0.28	10.14
Retentate 150 min	119.77	50.45	0.31	10.13
Retentate 180 min	119.77	50.41	0.29	10.13
Retentate 210 min	119.77	50.46	0.24	10.13
Retentate 240 min	119.31	50.46	0.33	10.15
Retentate 270 min	119.31	50.47	0.26	10.13
Retentate 300 min	119.31	50.43	0.35	10.14

Table C.40 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at original pH

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.046	0.046	1.00	0.039
5 min	0.046	0.046	1.00	0.039
10 min	0.046	0.046	1.00	0.039
15 min	0.046	0.046	1.00	0.039
30 min	0.046	0.044	0.95	0.039
45 min	0.046	0.040	0.87	0.039
60 min	0.046	0.037	0.80	0.028
75 min	0.046	0.034	0.73	0.028
90 min	0.046	0.031	0.67	0.028
105 min	0.046	0.027	0.59	0.021
120 min	0.046	0.023	0.50	0.021
135 min	0.046	0.021	0.45	0.014
150 min	0.046	0.019	0.40	0.014
165 min	0.046	0.016	0.35	0.014
180 min	0.046	0.014	0.30	0.007
195 min	0.046	0.012	0.25	0.18
210 min	0.046	0.009	0.20	0.09
225 min	0.046	0.008	0.17	0.09
240 min	0.046	0.007	0.15	0.09
255 min	0.046	0.006	0.14	0.09
270 min	0.046	0.006	0.13	0.09
285 min	0.046	0.006	0.12	0.09
300 min	0.046	0.005	0.11	0.09

Table C.41 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at original pH

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.13		50.57	0.25	7.87
Initial FM 30 min	103.68		50.74	0.67	8.07
Initial FM 60 min	83.54		40.67	0.92	8.02
Initial FM 90 min	69.96		40.54	0.59	8.05
Initial FM 120 min	68.65		40.96	0.73	8.04
Initial FM 150 min	66.46		40.65	0.65	8.06
Initial FM 180 min	65.59		40.87	0.54	8.05
Initial FM 210 min	63.84		40.95	0.85	8.07
Initial FM 240 min	63.40		40.74	0.77	8.05
Initial FM 270 min	62.52		40.89	0.91	8.04
Initial FM 300 min	62.08		40.56	0.83	8.05
Permeate 30 min	98.86	20.99	50.94	0.83	8.07
Permeate 60 min	80.91	35.34	40.63	0.97	8.07
Permeate 90 min	68.21	45.49	40.79	0.72	8.08
Permeate 120 min	67.78	45.84	40.87	0.57	8.06
Permeate 150 min	66.02	47.24	40.99	0.89	8.07
Permeate 180 min	65.15	47.94	40.61	0.44	8.03
Permeate 210 min	63.40	49.34	40.87	0.78	8.04
Permeate 240 min	62.96	49.69	40.76	0.55	8.03
Permeate 270 min	62.08	50.38	40.69	0.39	8.05
Permeate 300 min	61.65	50.73	40.92	0.64	8.07

Table C.42 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at original pH

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	110.25	50.71	21.8	7.48
Retentate 60 min	90.11	40.68	22.4	7.48
Retentate 90 min	81.35	40.85	21.7	7.47
Retentate 120 min	80.91	40.76	22.5	7.48
Retentate 150 min	72.15	40.83	22.9	7.47
Retentate 180 min	71.28	40.77	22.6	7.47
Retentate 210 min	71.72	40.98	21.2	7.48
Retentate 240 min	72.15	40.75	22.5	7.47
Retentate 270 min	71.72	40.68	22.8	7.47
Retentate 300 min	71.72	40.82	21.9	7.48

Table C.43 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at pH 4

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.047	0.046	1.00	1.00
5 min	0.047	0.046	1.00	1.00
10 min	0.047	0.046	1.00	1.00
15 min	0.047	0.046	1.00	1.00
30 min	0.047	0.044	0.95	1.00
45 min	0.047	0.041	0.89	0.92
60 min	0.047	0.037	0.81	0.82
75 min	0.046	0.034	0.74	0.73
90 min	0.046	0.031	0.67	0.64
105 min	0.046	0.028	0.60	0.64
120 min	0.046	0.025	0.53	0.46
135 min	0.046	0.022	0.47	0.46
150 min	0.046	0.019	0.41	0.37
165 min	0.046	0.016	0.35	0.28
180 min	0.046	0.014	0.29	0.28
195 min	0.046	0.012	0.26	0.18
210 min	0.046	0.011	0.23	0.18
225 min	0.046	0.009	0.20	0.09
240 min	0.046	0.008	0.17	0.09
255 min	0.046	0.007	0.15	0.09
270 min	0.046	0.006	0.14	0.09
285 min	0.046	0.006	0.13	0.09
300 min	0.046	0.005	0.12	0.09

Table C.44 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at pH 4

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	125.09		50.61	0.22	4.05
Initial FM 30 min	103.89		50.67	0.31	4.24
Initial FM 60 min	83.57		40.82	0.57	4.23
Initial FM 90 min	69.88		40.81	0.84	4.26
Initial FM 120 min	66.34		40.99	0.69	4.25
Initial FM 150 min	64.58		40.76	0.93	4.27
Initial FM 180 min	63.69		40.94	0.77	4.23
Initial FM 210 min	63.25		40.62	0.91	4.24
Initial FM 240 min	62.37		40.75	0.83	4.25
Initial FM 270 min	61.93		40.91	0.49	4.27
Initial FM 300 min	61.04		40.84	0.44	4.28
Permeate 30 min	99.03	20.83	50.48	0.72	4.27
Permeate 60 min	80.48	35.66	40.55	0.57	4.26
Permeate 90 min	68.11	45.55	40.62	0.89	4.28
Permeate 120 min	65.46	47.67	40.84	0.44	4.25
Permeate 150 min	64.13	48.73	40.73	0.78	4.27
Permeate 180 min	63.25	49.44	40.97	0.85	4.23
Permeate 210 min	62.81	49.79	40.54	0.57	4.24
Permeate 240 min	61.93	50.49	40.68	0.65	4.23
Permeate 270 min	61.48	50.85	40.74	0.54	4.22
Permeate 300 min	60.60	51.55	40.83	0.85	4.27

Table C.45 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at pH 4

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	110.51	50.68	22.7	3.87
Retentate 60 min	91.08	40.81	22.3	3.88
Retentate 90 min	80.92	40.94	21.2	3.88
Retentate 120 min	80.48	40.58	22.6	3.88
Retentate 150 min	71.64	40.77	21.1	3.88
Retentate 180 min	70.76	40.62	22.8	3.85
Retentate 210 min	70.32	40.59	22.4	3.88
Retentate 240 min	70.32	40.72	22.9	3.85
Retentate 270 min	70.32	40.91	22.5	3.89
Retentate 300 min	70.32	40.80	21.7	3.88

Table C.46 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at pH 7

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.047	0.047	1.00	0.039
5 min	0.047	0.047	1.00	0.039
10 min	0.047	0.047	1.00	0.039
15 min	0.047	0.047	1.00	0.039
30 min	0.047	0.044	0.93	0.039
45 min	0.047	0.041	0.87	0.035
60 min	0.047	0.037	0.79	0.035
75 min	0.045	0.034	0.73	0.025
90 min	0.045	0.031	0.65	0.025
105 min	0.045	0.028	0.58	0.017
120 min	0.045	0.025	0.52	0.017
135 min	0.045	0.022	0.46	0.014
150 min	0.045	0.019	0.40	0.014
165 min	0.045	0.016	0.34	0.014
180 min	0.045	0.014	0.29	0.007
195 min	0.045	0.012	0.25	0.007
210 min	0.045	0.010	0.22	0.004
225 min	0.045	0.009	0.20	0.004
240 min	0.045	0.008	0.17	0.004
255 min	0.045	0.007	0.15	0.004
270 min	0.045	0.006	0.14	0.004
285 min	0.045	0.006	0.13	0.004
300 min	0.045	0.005	0.11	0.004

Table C.47 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at pH 7

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	120.25		49.63	0.27	7.07
Initial FM 30 min	99.82		50.74	0.68	7.24
Initial FM 60 min	81.17		40.67	0.54	7.29
Initial FM 90 min	67.85		40.54	0.62	7.25
Initial FM 120 min	64.30		40.96	0.77	7.26
Initial FM 150 min	63.41		40.65	0.87	7.27
Initial FM 180 min	61.19		40.87	0.57	7.23
Initial FM 210 min	60.75		40.95	0.84	7.27
Initial FM 240 min	60.30		40.74	0.69	7.23
Initial FM 270 min	59.86		40.89	0.93	7.28
Initial FM 300 min	59.41		40.56	0.77	7.27
Permeate 30 min	94.94	21.05	50.96	0.74	7.21
Permeate 60 min	78.06	35.08	40.63	0.63	7.24
Permeate 90 min	66.07	45.05	40.87	0.72	7.22
Permeate 120 min	63.41	47.27	40.74	0.98	7.22
Permeate 150 min	62.97	47.64	40.55	0.85	7.29
Permeate 180 min	60.75	49.48	40.81	0.89	8.27
Permeate 210 min	60.30	49.85	40.73	0.78	7.28
Permeate 240 min	59.86	50.22	40.95	0.89	7.24
Permeate 270 min	59.41	50.59	40.77	0.92	7.25
Permeate 300 min	58.97	50.96	40.58	0.59	7.23

Table C.48 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at pH 7

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	106.93	50.83	21.3	6.77
Retentate 60 min	90.05	40.71	21.5	6.79
Retentate 90 min	80.28	40.64	21.4	6.78
Retentate 120 min	80.28	40.85	21.2	6.78
Retentate 150 min	71.40	40.73	21.6	6.79
Retentate 180 min	71.40	40.98	21.9	6.78
Retentate 210 min	71.40	40.58	21.4	6.77
Retentate 240 min	71.40	40.68	21.6	6.78
Retentate 270 min	70.52	40.83	21.8	6.78
Retentate 300 min	70.52	40.89	21.7	6.78

Table C.49 Permeate and retentate flux of formaldehyde solution by chitosan-enhanced ultrafiltration membrane at pH 10

Time	Water flux (M^3/M^2H)	Permeate flux (M^3/M^2H)	Normalized flux (M^3/M^2H)	Retentate flux (M^3/M^2H)
0 min	0.047	0.045	1.00	0.039
5 min	0.047	0.045	1.00	0.039
10 min	0.047	0.045	1.00	0.039
15 min	0.047	0.045	1.00	0.039
30 min	0.047	0.042	0.94	0.035
45 min	0.047	0.040	0.89	0.035
60 min	0.047	0.038	0.84	0.035
75 min	0.045	0.034	0.76	0.028
90 min	0.045	0.031	0.68	0.025
105 min	0.045	0.027	0.60	0.025
120 min	0.045	0.024	0.54	0.018
135 min	0.045	0.022	0.48	0.014
150 min	0.045	0.019	0.42	0.014
165 min	0.045	0.016	0.36	0.014
180 min	0.045	0.014	0.30	0.007
195 min	0.045	0.011	0.25	0.004
210 min	0.045	0.009	0.20	0.004
225 min	0.045	0.008	0.17	0.004
240 min	0.045	0.007	0.15	0.004
255 min	0.045	0.006	0.14	0.004
270 min	0.045	0.006	0.13	0.004
285 min	0.045	0.005	0.12	0.004
300 min	0.045	0.005	0.11	0.004

Table C.50 Formaldehyde concentration, percent formaldehyde removal, DOC, turbidity, and pH in feed and permeate by chitosan-enhanced ultrafiltration membrane at pH 10

	FM Conc. (mg/L)	%Removal	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Initial FM	120.02		49.53	0.26	10.05
Initial FM 30 min	100.35		50.43	0.74	9.84
Initial FM 60 min	80.24		40.81	0.63	9.82
Initial FM 90 min	67.57		40.99	0.59	9.83
Initial FM 120 min	64.07		40.76	0.98	9.85
Initial FM 150 min	63.20		40.94	0.85	9.87
Initial FM 180 min	61.89		40.62	0.68	9.85
Initial FM 210 min	61.01		40.75	0.54	9.86
Initial FM 240 min	60.58		40.59	0.62	9.85
Initial FM 270 min	60.14		40.73	0.77	9.84
Initial FM 300 min	59.27		40.87	0.87	9.84
Permeate 30 min	95.54	20.39	50.89	0.87	9.83
Permeate 60 min	77.19	35.69	40.78	0.95	9.87
Permeate 90 min	65.82	45.16	40.67	0.73	9.88
Permeate 120 min	63.20	47.34	40.92	0.48	9.83
Permeate 150 min	62.76	47.71	40.59	0.61	9.87
Permeate 180 min	61.45	48.80	40.73	0.59	9.86
Permeate 210 min	60.58	49.53	40.65	0.78	9.85
Permeate 240 min	60.14	49.89	40.54	0.97	9.85
Permeate 270 min	59.70	50.25	40.82	0.55	9.87
Permeate 300 min	58.83	50.98	40.48	0.72	9.84

Table C.51 Formaldehyde concentration, DOC, turbidity, and pH in retentate by chitosan-enhanced ultrafiltration membrane at pH 10

	FM Conc. (mg/L)	DOC (mg Carbon/L)	Turbidity (NTU)	pH
Retentate 30 min	106.03	50.79	21.8	9.65
Retentate 60 min	90.30	40.93	20.6	9.69
Retentate 90 min	80.24	40.85	20.8	9.68
Retentate 120 min	79.81	40.77	21.4	9.67
Retentate 150 min	71.07	40.53	21.7	9.65
Retentate 180 min	70.63	40.72	21.3	9.69
Retentate 210 min	70.19	40.65	21.9	9.67
Retentate 240 min	71.07	40.84	21.2	9.64
Retentate 270 min	70.63	40.51	21.7	9.68
Retentate 300 min	70.19	40.87	21.9	9.62

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