REDUCTION OF BACTERIOPHAGE IN SURFACE WATER BY COAGULATION WITH CERAMIC MEMBRANE MICROFILTRATION

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A Thesis Submitted in Partial Fulfillment of the Requirements
for the Degree of Master of Science Program in Environmental Management
(Interdisciplinary Program)
Graduate School
Chulalongkorn University
Academic Year 2013

การลดแบคทีรีโอฟาจในน้ำผิวดินโดยการรวมตะกอนร่วมกับไมโครฟิลเตรชันเซรามิกเมมเบรน

นางสาวปวีณ์ธิดา ยันต์ทองอยู่

วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิทยาศาสตรมหาบัณฑิต
สาขาวิชาการจัดการสิ่งแวดล้อม (สหสาขาวิชา)
บัณฑิตวิทยาลัย จุฬาลงกรณ์มหาวิทยาลัย
ปีการศึกษา 2556

Thesis Title	REDUCTION OF BACTERIOPHAGE IN SURFACE WATER BY COAGULATION WITH CERAMIC MEMBRANE MICROFILTRATION
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Field of Study	Environmental Management
Thesis Advisor	Associate Professor Suraphong Wattanachira, D.Eng.

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ปวีณ์ธิดา ยันต์ทองอยู่ : การลดแบคทีริโอฟาจในน้ำผิวดินโดยการรวมตะกอนร่วมกับไมโครฟิลเตรชันเซรามิกเมมเบรน. (REDUCTION OF BACTERIOPHAGE IN SURFACE WATER BY COAGULATION WITH CERAMIC MEMBRANE MICROFILTRATION) อ.ที่ปรึกษาวิทยานิพนธ์หลัก: รศ. ดร.สุรพงษ์ วัฒนะจีระ, 127 หน้า.

งานวิจัยนี้มีวัตถุประสงค์หลักเพื่อศึกษาการลดแบคทีริโอฟาจในน้ำผิวดินโดยการรวมตะกอนร่วมกับไมโครฟิลเตรชันเซรามิคเมมเบรน โดยศึกษาผลกระทบของขนาดรูพรุนของเซรามิกเมมเบรน (0.1, 0.5 และ1.0 μm), ปริมาณสารรวมตะกอน(1.5, 2.0, 2.5 และ3.0 mg-Al/L), และ ปริมาณความเข้มข้นเริ่มต้นของแบคทีริโอฟาจ (5.00E+05, 8.00E+06 และ 8.00E+07 PFU/mL) แบคทีริโอฟาจ Qβ เป็นตัวบ่งชี้ของไวรัสในลำไส้ของมนุษย์ซึ่งเป็นสาเหตุสำคัญของโรคที่เกิดจากน้ำเสีย ประสิทธิภาพการกำจัดของ แบคทีริโอฟาจ Qβ ใช้วิธีการ overlay plaque assay และรายงานผลตาม plaque forming unit (PFU) method ในการศึกษานี้ได้ทำการเก็บน้ำตัวอย่างจากแม่น้ำปิงจังหวัดเชียงใหม่ ประเทศไทย ในเดือนธันวาคม พ.ศ. 2554 ซึ่งมีค่าความขุ่นเท่ากับ 41.77 NTU และใช้ PACl (Polyaluminium Chloride) เป็นสารรวมตะกอน จากผลการศึกษาพบว่าขนาดรูพรุนของเซรามิกเมมเบรนที่มีขนาดเล็กมีประสิทธิภาพในลดแบคทีริโอฟาจ Qβ สูงกว่ารูพรุนขนาดใหญ่ เมื่อมีการประยุกต์ใช้ร่วมกับกระบวนการรวมตะกอนพบว่า ปริมาณของ PACl ที่ใช้ส่งผลกระทบต่อการลดแบคทีริโอฟาจ Qβ โดยค่าการกำจัดแบคทีริโอฟาจ Qβ สูงสุดเท่ากับ 7.9 log เมื่อใช้ไมโครฟิลเตรชันเซรามิกเมมเบรนขนาดรูพรุน 0.1 μm ร่วมกับการรวมตะกอนโดยใช้ PACl ปริมาณ 3.0 mg-Al/l การลดแบคทีริโอฟาจมีค่าต่ำสุดเท่ากับ 0.2 log เมื่อใช้ไมโครฟิลเตรชันเซรามิกเมมเบรนเพียงอย่างเดียว จากการศึกษาความเข้มข้นเริ่มต้นของแบคทีรีโอฟาจ Qβ พบว่า ถ้าความเข้มข้นเริ่มต้นของแบคทีรีโอฟาจสูง (8.00E+07 PFU/mL) มีผลทำให้การลดแบคทีริโอฟาจมีประสิทธิภาพลดลง จึงต้องใช้ปริมาณ PACl ให้มากขึ้นเพื่อให้สามารถรวมตะกอนได้มีขนาดใหญ่พอที่จะสามารถกำจัดได้โดยใช้ไมโครฟิลเตรชันเซรามิกเมมเบรนขนาดรูพรุน 0.1 μm เมื่อใช้ ปริมาณ PACl สูงสุดในการรวมตะกอน (3.0 mg-Al/l) ไมโครฟิลเตรชันเซรามิกเมมเบรนขนาด 0.5 μm และ 0.1 μm จะมีความสามารถในการกำจัด แบคทีรีโอฟาจ Qβ เท่ากัน ดังนั้นการใช้การรวมตะกอนด้วย PACl ร่วมกับ 0.5 μm ไมโครฟิลเตรชันเซรามิกเมมเบรนจึงเป็นสภาวะที่เหมาะสมที่สุดในการกำจัดแบคทีรีโอฟาจ Qβ เนื่องจากสามารถผลิตน้ำได้ในปริมาณที่มากกว่า

สาขาวิชา การจัดการสิ่งแวดล้อม ลายมือชื่อนิสิต .....

ปีการศึกษา 2556 ลายมือชื่อ อ.ที่ปรึกษาวิทยานิพนธ์หลัก .....

5387546420 : MAJOR ENVIRONMENTAL MANAGEMENT
KEYWORDS: CERAMIC MEMBRANE / BACTERIOPHAGE / PACL / IN-LINE COAGULATION

PAVEETIDA YANTHONGYU: REDUCTION OF BACTERIOPHAGE IN SURFACE WATER BY COAGULATION WITH CERAMIC MEMBRANE MICROFILTRATION. ADVISOR: ASSOC. PROF. SURAPHONG WATTANACHIRA, D.Eng., 127 pp.

The main objective of this study was examining the reduction of F-specific RNA bacteriophage Qβ from spiked-surface water by coagulation with ceramic membrane microfiltration. The effects of pore size of the ceramic membrane (0.1, 0.5 and 1.0 μm), coagulant dosages (1.5, 2.0, 2.5 and 3.0 mg-Al/L), and initial bacteriophage Qβ concentration (5.00E+05, 8.00E+06 และ 8.00E+07 PFU/mL) to the reduction of F-specific RNA bacteriophage Qβ were investigated. Bacteriophage Qβ was used as an indicator of human enteric viruses which a major cause of waterborne diseases. The reduction performance of Qβ was measured by overlay plaque assay and reported in plaque forming unit (PFU) method. In this study, water sample was collected from Ping River in December 2011 (Chiang Mai, Thailand), which contained turbidity of 41.77 NTU. Polyaluminium Chloride (PACl) was used as a coagulant in coagulation process. From the experiment results, the smaller pore size ceramic membrane, microfiltration yields higher bacteriophage Qβ log removal. When the coagulation was applied, coagulants dosage strongly affected bacteriophage Qβ removal. The high log removal (7.9) was achieved with 0.1 μm ceramic membrane pore size at 3.0 mg-Al/L PACl dosage, while 0.2 log removal was observed by ceramic membrane microfiltration alone. Furthermore bacteriophage Qβ concentrations in feed water affected the removal efficiency as well. The high initial Qβ concentration (8.00E+07 PFU/mL) was affected the reduction efficiency. It was required more amount of PACl dosage to form the large aggregate which larger than the pore size of 0.1 μm ceramic membrane microfiltration. At the highest PACl dosage coagulation(3.0 mg-Al/l) , 0.5 μm and 0.1 μm pore size achieved equivalent capability to reduce bacteriophage Qβ. Thus, the PACl coagulation with 0.5 μm ceramic membrane filtration was the achievable condition for reduce bacteriophage Qβ since it can produce in larger filtrated volume than 0.1 μm.

Field of Study: Environmental Management Student's Signature .....

Academic Year: 2013 Advisor's Signature .....

ACKNOWLEDGEMENTS

I would like to express my sincere gratitude to my thesis advisor, Associate Professor Dr. Suraphong Wattanachira at the Department of Environmental Engineering of Chiang Mai University for his kindness, valuable suggestions, guidance, advice, and especially strong encouragement throughout this research.

Special respect and thanks are also extended to Dr. Aunnop Wongrueng at the Department of Environmental Engineering of Chiang Mai University for his valuable suggestion and guidance.

I would like to sincerely thank the National Research Center for Environmental and Hazardous Waste Management (NRC-EHWM), Chulalongkorn University for the financial support for this research work.

Special thanks are also made to all students and staff at the National Center of Excellence for Environmental and Hazardous Waste Management (NCE-EHMW) program, Chulalongkorn University. Especially Parkphum Rakruam, for all his help during the experimentation and dissertation writing processes, valuable suggestion and guidance. I would like to express gratitude to all staff members of the Department of Environmental Engineering of Chiang Mai University for the use of their laboratory facilities and providing valuable information. I also thank all friends at the Department of Environmental Engineering, Chiang Mai University for their support and helps over the entire experimental period.

Finally, I could not accomplish without the inspiration from my family throughout my research work even on the difficult time.

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LIST OF ABBREVIATIONS

AMW	Apparent Molecular Weight
AWWA	American Water Works Association
CaCO3	Calcium Carbonate
CM	Ceramic membrane
CHBr3	Bromoform
CHCl2Br	Bromodichloromethane
CHCl3	Chloroform
CHClBr2	Dibromochloromethane
Cl	Chlorine
cm	Centimeter
°C	Degree Celsius
DOC	Dissolved Organic Carbon
DOM	Dissolved Organic Matter
EPA	Environmental Protection Agency
g/cm3	Gram per Cubic Centimeter
g/L	Gram per Liter
g/mol	Gram per Molar
hr	Hour
L/mg-m	Liter per milligram-meter
MF	Microfiltration
MCL	Maximum Contaminant Level
mg/l	Milligram per Liter
MWCO	Molecular Weight Cut Off
NOM	Natural Organic Matter
NTU	Nepheo Turbidity Unit

PACl	Polyaluminium Chloride
S	Second
SUVA	Specific Ultraviolet Absorption
THMFP	Trihalomethanes Formation Potential
THMs	Trihalomethanes
TOC	Total Organic Carbon
UF	Ultrafiltration
USEPA	United States Environmental Protection Agency
UV-254	Ultraviolet absorption at wave length 254 nanometer
UV	Ultraviolet Absorption

CHAPTER I

INTRODUCTION

1.1 Motivations

Membrane filtration technology for municipal water and wastewater treatment has been used extensively all over the world, and the installation of microfiltration (MF) and ultrafiltration (UF) facilities has dramatically increased over the past decade (Adham *et al*., 2005). Membrane can be observed as an absolute barrier to the different types of contaminant that will be physically larger than the largest pore on the membrane. MF and UF are considered as a capable process to provide better quality drinking water for the water supply. Membrane applications are receiving increased attention associated with water quality and cost reduction by improvements in membrane technology (Sangyoup *et al*., 2004).

The use of microfiltration and ultrafiltration for water and wastewater treatment has been almost exclusively focused on polymeric membranes (Van der Bruggen *et al*., 2003). Most polymeric membrane systems operate rightly within a fairly neutral to slightly acidic pH range, with extended extreme acidic or alkaline conditions posing potential problems (Farahbakhsh *et al*., 2004). In addition, exposure to extreme oxidant conditions created by chlorine and ozone can cause degradation of polymeric membranes (Castro *et al*., 1995). As a result, innovative MF/UF membranes are presently being developed to improve pore distribution,

mechanical stability, and chemical stability, using optimized polymeric formulations or alternative materials (i.e., ceramics, steel, polytetrafluoroethylene, etc.).

Ceramic membranes are well known in the current challenges of conventional polymeric membranes that have the combined advantages of high mechanical strength, stability for chemical, high permeability, thermal resistance and long service life. Moreover, the ceramic membranes exhibited higher permeability than the equivalent polymeric membranes (Lee S. *et al*., 2004). Ceramic membrane pore size covers the MF, UF and tight UF ranges from 5 μm down to 1000 Daltons of molecular weight cut-off (MWO) (Sondhi *et al*., 2003). These unique thermal, chemical and mechanical properties of ceramic membrane have significant advantages over polymeric membranes in many applications.

The ceramic membranes are used to remove particular matter such as inorganic particles as well as microorganisms including bacteria and viruses. Nevertheless, ceramic membrane is MF and UF whose pore sizes are not small enough to reject particles smaller than tens of nanometers and thus cannot remove viruses that cause health concerns from water (Matsushita *et al*., 2005). Virus removal could be enhanced through mechanical sieving by membrane or adsorption onto the membrane, as well as by cake layer during MF filtration (Jacangelo *et al*., 1995). Hence a hybrid membrane system using coagulation or adsorption is required to enhance virus removal of MF membrane alone (Matsushita *et al*., 2004). Therefore, the coagulation and flocculation processes that have been previously introduced to enhance virus aggregate formation prior to membrane filtration process. Thus, they

are expected to have large advantage to combine with coagulant adsorption for controlling membrane fouling and improving removal rates.

The combination of ceramic membrane filtration and coagulation process was developed to increase the efficiency of microorganism or DOM removal from surface water. Generally, the removal mechanism of ceramic membrane is size separation. However, the lower pressure membrane such as microfiltration has larger pore size than microorganism and DOM. Then, the removal efficiency was limited. The coagulation process was applied to increase the removal efficiency by using the aggregation mechanism with coagulants such as adsorption, entrapment and charge neutralization (Jarvis *et al*., 2004). The addition of coagulants during the coagulation process can be increased the size of aggregates to have larger than the membrane pore size (Matsushita *et al*., 2005). In addition, many researchers were investigated that the better performance and filtrated water quality of coagulation with UF membrane depended on the good coagulation condition including coagulant type, dose and pH (Guigui *et al.*, 2002).

Coagulation process is a chemical reaction which is uses to remove suspended matter. The coagulant encourages colloidal material in the water to join together into small aggregates. Many researches showed the advantage of using coagulation process combined with membrane filtration. Polyaluminium chloride (PACl) is a class of coagulant which was developed in Japan and widely used for water treatment in Asia and Europe. Many researchers revealed that PACl coagulants have several advantages over the traditional metal salt coagulants such as higher

quality of the treated water, better overall purification efficiency and shorter flocculation time, wider working pH range and lower residual aluminum concentration.

Human enteric viruses have been recognized as a major cause of waterborne outbreaks which have been reported worldwide in both developed and developing countries (Hoebe *et al*., 2004; Boccia *et al*., 2002). Human enteric viruses can survive for extended times in the environment under a wide range of pH and temperatures. The numbers of enteric viruses presented in surface waters are generally few and difficult to identify and isolate. Therefore, the basic steps of virological analysis of environmental waters are consisted of sampling, virus concentration and detection.

From the study of Schijven and Hassanizadeh, Challenge testing of membrane processes with native viruses is not practically feasible because their concentrations are very low in water samples. Moreover assay of native viruses is complex and time consuming for some viruses definite analysis methodology is not available (Schijven and Hassanizadeh, 2000; Templeton *et al*., 2008). For that reason, challenging the membrane processes for virus removal is generally performed with model viruses under conditions where its inactivation and adsorption behaviors are similar to the native viruses under given conditions.

The human enteric virus analysis requires an advance technology, specialists, and its time consuming and inaccurate as they exit at low concentrations in the

environment. Therefore, Interest was focused on indicator organisms that are nonpathogenic, rapidly detected, easily enumerated, similar survival characteristics to those of the pathogens and able to associate with the presence of pathogenic microorganisms.

The bacteriophages (bacterial viruses) have been proposed as useful alternative viral indicator, as their morphology and survival characteristics closely resemble those of some of the important human virus groups. Several researches have been published on the use of bacteriophages as viral indicators for the presence of human enteric viruses in fresh water, indicators of fecal pollution of treated or untreated drinking water, or indicators of treatment efficiency. The advantage of using bacteriophages as a surrogate is that they can be prepared in large quantities and in high concentration for seeding in challenge studies, enabling demonstration of up to 11-log removal (Schijven and Hassanizadeh, 2000).

The F-specific RNA bacteriophage, Qβ (diameter 0.023 μm, pI 5.3) has been used extensively as a surrogate virus for waterborne viruses because of their morphological and structural resemblance to human enteric viruses (Matsui *et al*., 2003; Matsushita *et al*., 2005; Otaki *et al*., 1998; Shirasaki *et al*., 2007; Urase *et al*., 1996). In addition, its survival characteristics in aquatic environments are similar to those of human enteric viruses.

This research discussed results of lab-scale studies designed to investigate ceramic membrane filtration technology for natural water treatment, focusing on

bacteriophage Qβ removal efficiency by ceramic membrane filtration. Furthermore, the combination of PACl coagulation with ceramic membrane filtration will be demonstrated. In order to evaluate the filtrate water quality through the viral indicators, bacterial host strain named *E. coli* K12 and *Salmonella typhimurium* WG 49 were used for F-specific RNA bacteriophages enumeration.

1.2 Objectives:

The main objective of the study was to examine bacteriophage Qβ reduction efficiency by ceramic membrane filtration combined with coagulation. In order to achieve the main objective, the following sub objective should be considered.

1. To investigate the effect of bacteriophage Qβ concentration in surface-water to the efficiency of bacteriophage Qβ reduction by ceramic membrane filtration.
2. To determine the most achievable concentration of coagulants at different becteriophage Qβ concentration and different pore sizes of ceramic membrane.

1.3 Hypotheses:

1. Differences in membrane pore sizes of ceramic membranes affect bateriophage Qβ reduction efficiencies.

2. A combination of coagulation and ceramic membrane filtration can increase bacteriophage Qβ reduction efficiency.

3. Bacteriophage concentrations in water affect the bacteriophage Qβ reduction efficiencies.

1.4 Scopes of the Study:

1. Ping River, Chiang Mai, Thailand was used as raw surface water. The characteristics of raw surface water was analyzed by measuring various parameters including turbidity, pH, DOC, UV-254, Alkalinity and Temperature.

2. F-specific RNA bacteriophages (Qβ) were used as a model virus and detected to evaluate filtrate quality by overlay plaque assay using *Salmonella typhimurium* WG 49 as host strain.

3. Batch experiment of the ceramic membrane filtration was studied.
   - The reduction of bacteriophage Qβ concentration by ceramic membrane filtration was determined in log removal value.
   - The efficiency of ceramic membrane at different pore sizes was determined.

4. Polyaluminium Chloride (PACl) was used as coagulant for coagulation combined with ceramic membrane filtration at different pore sizes (1.0 μm, 0.5 μm and 0.1 μm).

5. Batch experiment of ceramic membrane filtration with coagulation was studied.
   - The efficiency of ceramic membrane with coagulation was determined.
   - PACl was varied to find the most achievable PACl dosage for different ceramic membrane pore sizes.
   - The most achievable PACl dosage for different ceramic membrane pore sizes was determined.

6. Batch experiment of inline coagulation at the most achievable PACl dosage with ceramic membrane filtration was studied.
   - The effect of bacteriophage concentrations in surface water affects the bacteriophage Qβ reduction efficiencies.
   - The most achievable PACl dosage of different becteriophage Qβ concentration and different pore sizes was investigated.

CHAPTER II

BACKGROUND AND LITERATURE REVIEWS

2.1 Membrane filtration

In recent years, applications of membranes filtration in drinking water treatment have shown a significant increase. Especially for surface water treatment, the membrane filtration has gained interest in the field of water supply or drinking water. The membranes are used to remove particulate matter such as inorganic particles as well as microorganisms including bacteria and virus over the conventional coagulation. In addition, Membrane filtration also have many advantages including require lower space to treat a given flow, require lower chemical consumption, easy to operate and maintenance and provided the better water quality (Nakatsuka *et al*., 1996). The retention of a virus is dependent on the type of membrane and membrane characteristic, module design and operating conditions.

Membrane filtration membranes, which are presently in operation in European waterworks, are made from organic material. Recent developments showed that membranes made from inorganic materials could be promising in membrane technology in the future, because of their unique characteristics including a hydrophilic surface and a high resistance against mechanical, chemical or thermal stress (Lerch *et al*. 2005, Heijman and Bakker 2007, NGK 2008, METAWATER 2009).

Furthermore, membrane filtration is ecologically friendly and more favorable than other separation technologies.

2.1.1 Ceramic Membrane Filtration Technology

Ceramic materials are stable chemically, thermally and mechanically. They are ecologically friendly and more favorable than other separation technologies. No additives are necessary and the process temperature is not limited. Filtration with ceramics is a mild and highly selective process without phase transformation. Running costs are limited by closed production cycles and continuous processes. They are therefore ideal materials for many applications in the chemical and pharmaceutical industries or in water and wastewater treatment. The membrane modules can withstand elevated temperature, extremes of pH (0 to 14), and high operating pressures up to 10 bar (145 psi). For these reasons, membrane compaction, delaminating or swelling, which makes these membranes suitable for many applications but polymeric and other inorganic membranes, cannot be used. Additionally, ceramic membranes are ideal for in-place chemical cleaning at high temperatures using caustic, chlorine, hydrogen peroxide, ozone and strong inorganic acids, as well as, steam sterilization. Limitations of filtration technology, the advantages and disadvantages of ceramic membrane technology are presented in Table 2.1 and Table 2.2.

Table 2.1 Limitations of filtration technology.

Filtration Technology	Limitation
Diatomaceous Earth	• Recovered product quality is poor. • Spent DE disposal problematic. • Blinding of media with contaminants yields low flux or productivity.
Polymeric Membrane	• No polymeric membrane available with the required submicron pore size and solvent stability.

Table 2.2 The advantages and disadvantages of ceramic membrane technology

Advantages	Disadvantages
1. Can be used to treat entire range of high flash solvents. 2. Excellent recovered product quality. 3. Low temperature operation. No thermal degradation of solvent. 4. Good product recovery ratios. 5. No additional waste disposal problem. 6. Technology is easily implemented. No special operator training required and minimal maintenance.	1. Cost of ceramic membrane is still high. 2. Broken easier 3. Range of molecular rejection was low compare to polymer membrane.

2.1.2 Materials

Ceramic membranes move across the range from A to Z depending on the conditions of the materials (from alpha alumina to zircon). The membranes are mostly made from Al, Si, Ti or Zr oxides. Ti and Si oxides are more stable than Al or Si oxides. In some infrequent cases, Sn or Hf is used as a base element. Each oxide has a common surface charge when presented in a solution. Some membranes consist of mixed oxides, which are established by some supplementary compounds presenting an insignificant concentration.

The supports for the membrane elements are made from γ-aluminium oxide or silicon carbide with open pores. This material can provide not only maximum permeability but also fulfill high requirements relating to mechanical stability. These supports are either for a single channel or a multi-channel design. A membrane layer of a define texture only a few micrometer (μm) thick is applied to the inner side of the channels in a sandwich-type process and connected monolithically.

2.1.3 Structure

Ceramic membranes are available in various pore sizes including microfiltration, ultrafiltration and tight ultrafiltration. Ceramic membranes show an asymmetrical structure, which consist of at least two layers (mostly three layers) with different porosity levels. Generally, there are two main layers combined in ceramic membranes i.e., separation layer and supporting layer, as illustrated in Figure 2.1.

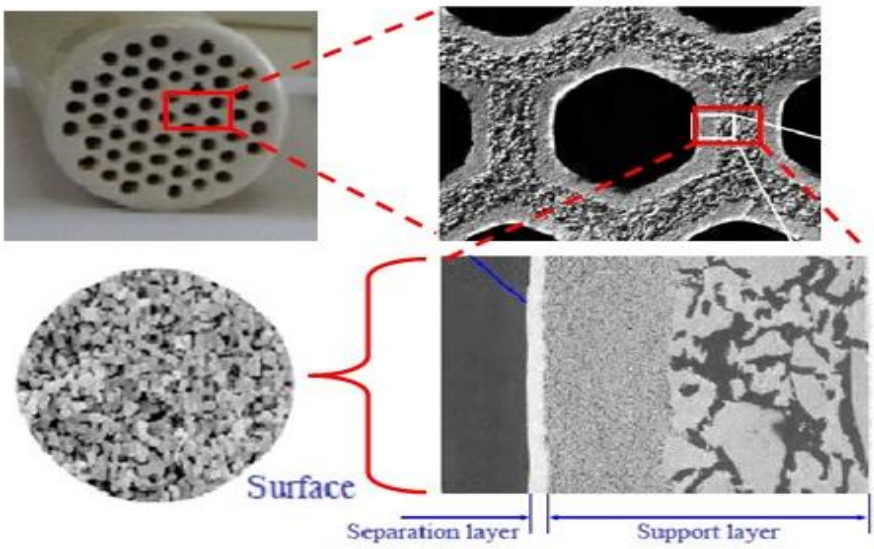


Figure 2.1 The magnification of ceramic membrane structure (Source: Meta Water Co., Ltd. Confidential Report, 2008)

The ceramic membranes are shaped in an asymmetric, multi-channel element. These elements are combined together in housings, and these membrane modules can endure high temperatures of extreme acidity or alkalinity and high operating pressures, making them suitable for many applications where polymeric and other inorganic membranes cannot be available. Several membrane pore sizes are provided for specific filtration requirement covering the microfiltration, the ultrafiltration, and nanofiltration ranges (from 5 μm down to 1000 Daltons).

2.1.4 Flow

Membrane filtration is a pressure driven process, which can remove impurities from a solution (so-called a feed solution) by a semi-permeable property of a membrane. A filtered solution through the membrane is defined as permeate or

filtrate. The standard ceramic filter elements have in common, which the membrane layer is fixed at the inside of the tubes.

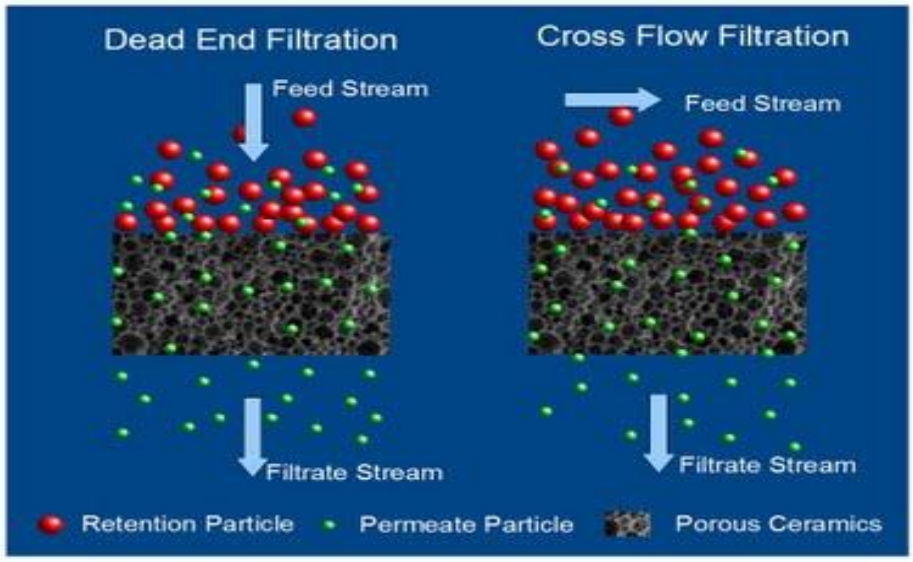


Figure 2.2 The mechanics of dead-end filtration and cross- flow filtration ("Porous Ceramic Application: Porous Ceramics Filtration & Separation Application" [Online]. Available from: http://www.induceramic.com, 2014)

Membrane filtration systems can be operated in 2 modes, i.e. cross-flow filtration and dead-end filtration (figure 2.2). Dead-end filtration is a system that can feed a solution in a direction perpendicular to a membrane. It is suitable for filtering a solution containing low impurities, which tend to clog the membrane easily. In addition, dead-end filtration is widely used as a laboratory-scale experiment. Cross-flow filtration is a system that feed a solution parallel to the membrane surface causing a shear force between the surface membrane and the feed solution. It can control a membrane fouling since it can sweep particles or impurities out of the surface membrane. Cross-flow filtration is suitable for using on an industrial-scale.

2.1.5 Element shapes

Ceramic membranes are available from several manufacturers in different shapes, mainly round and hexagonal, and with various channel diameters. A multichannel construction provides a higher membrane packing density than a tubular element of the same length. A typical industrial installation will have several of these modules arranged in series and/or parallel configuration.

Figure 2.3 Typical element designs on ceramic membrane ("Filtration membranes: membrane materials and pore sizes" [Online]. Available: http://www.rauschert.de, 2009)

The membrane elements have to be designed in a way, that they can handle also feed media with a high content of particles or a high viscosity. At last, the geometries of the ceramic filter elements are responsible that the hydraulic properties during the process are acceptable all over the membrane element. Therefore, the membrane elements are designed with tubular channels; depending on the application and properties like e.g. viscosity and particle content, they are used in single- channel design or in multi-channel design. The membrane elements

under discussion are hexagonal in 3 centimeter in diameter, 10 centimeters height and tubular channels. These are available with three different pore sizes (1.0 μm, 0.5 μm and 0.1 μm).

2.1.6 Applications

Ceramic membranes are being used increasingly in a broad range of industries such as biotechnology and pharmaceutical, dairy, food and beverage, as well as chemical and petrochemical, microelectronics, metal finishing, and power generation. Each industry presents specific needs and opportunities. The membrane modules can withstand elevated temperatures, extremes of pH (0 to 14), and high operating pressures up to 10 bar (145 psi) without concern for membrane compaction, delaminating or swelling. This makes these membranes suitable for many applications where polymeric and other inorganic membranes cannot be used. Additionally, ceramic membranes are ideal for in-place chemical cleaning at high temperatures, while using caustic, chlorine, hydrogen peroxide, ozone and strong inorganic acids, and/or by using steam sterilization.

2.2 The combination system of ceramic membrane and coagulation

The combination of ceramic membrane filtration and coagulation process was developed to increase the efficiency of microorganism or DOM removal from surface water. Generally, the removal mechanism of ceramic membrane is size separation. However, the lower pressure membrane such as microfiltration has larger pore size

than microorganism and DOM. Then, the removal efficiency was limited. Many researchers was studied the removal efficiency of DOM by using the combination of membrane filtration with coagulation process. It shows that the ceramic membrane filtration process with coagulation effected to removing suspended solids from several river water samples in Southeast Asia (Hata *et al*., 2009). The coagulation process was applied to increase the removal efficiency by using the aggregation mechanism with coagulants such as adsorption, entrapment and charge neutralization (Jarvis *et al*., 2004). The addition of coagulants during the coagulation process can be increased the size of aggregates to have larger than the membrane pore size (Matsushita *et al*., 2005). In addition, many researchers were investigated that the better performance and filtrated water quality of coagulation with UF membrane depended on the good coagulation condition including coagulant type, dose and pH (Guigui *et al.*, 2002).

2.3 Coagulation process

2.3.1 Conventional coagulation

Coagulation and flocculation are the adding of chemical reagent to destabilize of colloid particles which it can easier to combine together. Normally, the surface charges of colloid particles are negative which cannot combine together. Thus, the adding of chemical can be neutralized the surface charge of colloid particles which it easier to agglomerate. Coagulation referred to the addition of coagulants and rapid mixing which cause of destabilization of the colloid particles. Then, the destabilization colloid particles were agglomerated. The flocculation is the

slow mixing which the destabilization colloid particles can be aggregate to form floc. Subsequently, the floc was removed by sedimentation or filtration.

2.3.2 Mechanisms of coagulation

Four mechanisms can be used to explain the particle destabilization: (1) double layer compression, (2) adsorption or charge neutralization, (3) enmeshment in a precipitation, and (4) adsorption and antiparticle bridging. Normally, the coagulation is the process of particles charge destroyed. The mechanism which related was double layer compression and charge neutralization. While the enmeshment and bridging is related to flocculation process (Benefield *et al*., 1982).

2.3.2.1 Double Layer Compression

Double layer compression are involves the electrostatic repulsion. It occurs when the counter-ions is added as coagulants. The highest concentration of counter ions is found at the surface of particles and decreases at the outer boundary of diffusion layer. The compression of diffusion layer can lead the destabilized of particles by counter-ions. It can decrease the electrostatic repulsive forces between similar particles and the zeta potential is mitigated. Therefore, the particles are bind together with the attractive forces (van der Waals forces).

2.3.2.2 Charge Neutralization

Charge neutralization occurs when a colloid particle is destabilized by the coagulant ions. When the coagulants dissolves in water, the positive charged of

coagulants ions neutralizes the negative charge of colloid particles. Thus, the charge of particle is reduced to the level that particles are destabilized. Then, the colloid particles can be adsorbed together.

2.3.2.3 Sweep Coagulation

Sweep coagulation involves the formation of a solid precipitate. This mechanism occurs when the enough concentration of coagulants was added. The crystal of coagulants is covering the colloid particles. So, the negative charge of colloids particles is enmeshed to the precipitates.

2.3.2.4 Interparticle Bridging

Destabilized particles can be aggregated by bridging with a polymer. Interparticulate bridging refers to the interaction between the polymer and the reactive groups on the destabilized particles. When a high molecular weight polymer comes into contact with a colloidal particle, some of the reactive groups in the polymer adsorb at the particle surface and leaving other portions of the molecule extending into the solution (AWWA, 1990).

2.3.3. Factor influence coagulation process

2.3.3.1. Characteristics of natural organic matter (NOM)

Characteristics of NOM in water are depended on the origination and geology. Thus, NOM characteristics in various place or country are different which

affect the coagulation process. Kim and Yu (2005) and Sharp *et al*. (2006) reported that NOM which defined as hydrophobic were easier to remove than hydrophilic and the high molecular of NOM are higher remove by coagulation than small molecular of NOM.

2.3.3.2. Types and concentration of coagulants

There are many types of coagulants. The different types of coagulants provided the different ability to remove NOM in water. Many researchers investigated the performance of different coagulants for NOM removal. Uyak and Ismail (2007) studied the NOM removal by using AL^{3+} and Fe^{3+} and found that Fe^{3+} can remove NOM better than AL^{3+}. In addition, Musikavong (2005) studied the removal of NOM and THMFP by using alum and $FeCl_3$ and reported that both Alum and FeCl3 can remove NOM with percent removal 35% at coagulants concentration 40 mg/L.

2.3.3.3. pH

The variation of pH of water was found to affect the coagulation process. Many researcher including Kabsch-Korbutowicz (2005); Qin *et al*. (2006) and Uyak and Ismail (2007) were studied the effect of pH on the coagulation process and concluded that the different of pH was affected to the performance of coagulation process.

2.4 Membranes filtration for microbial removal

Microfiltration, typically with pore sizes 0.1 μm have shown lower removal of viruses, and in some cases, could not act as a physical barrier to viruses (Sondhi *et*

al., 2003). Ultrafiltration can achieve more than 6 log (99.9999%) virus removal; Microfiltration cannot efficiently remove viruses when the filtration mechanism relies on physical sieving alone. The addition of coagulant, the most commonly used methods for the removal of suspended solids in water, is one of the selections of the pretreatment process before a membrane filtration process to increase permeate quality.

Several studies have reported the usefulness of the coagulation process for the removal of enteric viruses and bacteriophages, which are viruses that infect bacteria (Guy *et al.,* 1977; Havelaar *et al.,* 1995). Previous researches have been presented that some viruses have a tendency to adsorb on to the aluminum floc particles, which are finally retained by the membrane to form the cake layer (Clesceri *et al*., 1998).

Other researchers have reported that the formation of a cake layer may enhance the removal of viruses by membrane filtration (Jacangelo *et al*., 1995; Madaeni *et al*., 1995; Farahbakhsh *et al*., 2004) because the PACl accumulated in the membrane compartment would consequently increase with time and could inactivate the viruses there (Matsushita *et al*., 2005). Complete removal of poliovirus (Madaeni *et al*., 1995) and more than 6 logs of MS2 virus removal (Jacangelo *et al*., 1995) was obtained using 30 and 100 kDa molecular cut off membranes, respectively, whereas incomplete Qβ virus retention (2.5 logs) has been observed using UF membranes in the 30 kDa range (Urase *et al*., 1996). In addition, the removal ratio of the infectious Qβ concentration was approximately 2 log higher than the infectious

MS2 concentration of all coagulant dosages tested. Qβ was more sensitive to the virucidal activity of the aluminum coagulant (Shirasaki *et al*., 2009)

Matsui *et al*., 2005 concluded that the coagulant dose, membrane pore size, and coagulation time affected virus removal. Increasing the coagulant dosage was most effective for virus removal. Extending time probably improved the low removal resulting from the low coagulant dose. The effect of membrane pore size was more clearly observed at the beginning of filtration where the caked layers have not fully developed. Microfiltration with nominal pore size of 0.1μm after coagulation pretreatment with the PACl dose of 1.08 mg/l Al and 2.4-s mixing time was achieved over a 6.4 log reduction in virus load. The microfiltration whose pore sizes were 0.5 and 0.1 μm showed about 1 log less removal than by the 0.1 μm pore-size.

2.5 Polyaluminium Chloride (PACl) coagulation

For the duration of drinking water treatment, coagulation is an essential process for combining small particles into larger aggregates. Small particles in the drinking water source, such as viruses, that will not settle from suspension by gravity are destabilized and combined into larger aggregates during the coagulation process. In a conventional coagulation-sedimentation process, sufficient mixing time is required so that coagulation and flocculation occur and the aggregates grow large enough to settle down under gravity. Nevertheless, long-duration mixing is probably not needed in the coagulation-MF hybrid system (Judd and Hillis., 2001).

The term "poly-aluminum chloride" or "PACl" refers to a class of soluble aluminum products in which aluminum chloride has been partly reacted with a base. The relative amount of OH-, compared to the amount of Al, determines the basicity of a particular PACl product. The chemistry of PACl is often expressed in the form $Al_n(OH)_mCl_{3n-m}$. Solutions of PACl are not as acidic as alum; consequently they do not tend to decrease the pH as much as an equivalent amount of alum. Another difference is that PACl is formulated so that it already contains some of the highly cationic oligomers of aluminum - materials that are especially effective for the modification of colloidal charges. A particularly stable and important ionic species in PACl and related soluble aluminum chemicals has the formula Al_{12} $(OH)_{24}AlO_4$ $(H_2O)_{12}^{7+}$. Basicity can be defined by the term m/(3n) in that equation.

Polyaluminium chloride (PACl) is increasingly used for water treatment. Against the conventional use of aluminium sulphate (alum), it has shown distinct advantages. PACl are synthetic polymers dissolved in water. They react to form insoluble aluminium poly-hydroxides which precipitate in big volumetric flocs. The flocs absorb suspended pollutants in the water which are precipitated with the PACl and can together be easily removed. PACl can be used as a flocculants for all types of water treatment, drinking water, industrial waste water, urban waste water and in the paper industry. Chaimongkol (2008) concluded that the advantages of PACl over Alum were,

- Lower dosage requirement
- No requirement for any neutralizing agent (soda, lime)
- Shorter flocculation time

- Smaller amount of sludge
- Reduced number of back washing steps
- Higher quality of treated water
- Alum can contains many type of hazardous metals in some conditions

2.6 Removal of organic matter by coagulation process

Coagulation process is utilized in water supply process. It can remove both turbidity and dissolved organic matter. Many researchers utilize coagulation process with PACl to remove DOM. Rizzo (2005) studied the efficiency of alum, PACl and $FeCl_3$ for NOM reduction. The results showed that the using PACl as coagulants can remove turbidity with highest percent removal compared to alum and $FeCl_3$. Zhonglian *et al*. (2010) used the coagulation process with alum and PACl to remove NOM from surface water. The results showed that the using of PACl as coagulants provided the higher percent NOM removal than using alum. The percent removal of turbidity, DOC and UV-254 of coagulation with PACl were 94.5%, 34.8% and 53.5%, respectively. Furthermore, it was found that the using of PACl as coagulants has the residual aluminium after treatment lower than using of alum.

2.7 Viral Indicators

Pathogens are biological agents that are capable of causing disease or illness to its host. The major pathogens of concern include viruses, bacteria and protozoa. A list of the important waterborne pathogens which have there is evidence to their

occurrence in drinking water supplies, given by the World Health Organization (WHO) guidelines (Table 2.3). The waterborne in the list were show the significant effect and has been confirmed by epidemiological studied and case history. The most significant virus groups affecting water quality and human health originates in the gastrointestinal tract of infected individuals, called human enteric viruses.

Table 2.3 Waterborne pathogens of concern and their significance in water supplies (Antony *et al*., 2012)

Pathogen	Health significance	Persistence in water supplies[a]	Resistance to chlorine[b]	Relative infectivity[c]	Important animal source
Viruses					
Enteroviruses	High	Long	Moderate	High	No
Astroviruses	High	Long	Moderate	High	No
Hepatitis A viruses	High	Long	Moderate	High	No
Hepatitis E viruses	High	Long	Moderate	High	Potentially
Noroviruses	High	Long	Moderate	High	Potentially
Rotavirus	High	Long	Moderate	High	No
Bacteria					
Burkholderia pseudomallei	High	May multiply	Low	Low	No
Escherichia coli–Pathogenicd	High	Moderate	Low	Low	Yes
E. coli–Enterohaemorrhagic	High	Moderate	Low	High	Yes
Legionella spp.	High	May multiply	Low	Moderate	No
*Pseudomonas aeruginosa*e	Moderate	May multiply	Moderate	Low	No
Salmonella typhi	High	Moderate	Low	Low	No
Other salmonellae	High	May multiply	Low	Low	Yes
Shigella spp.	High	Short	Low	High	No

TABLE 2.3. Waterborne pathogens of concern and their significance in water supplies (Antony *et al*., 2012). (countinue)

Pathogen	Health significance	Persistence in water supplies[a]	Resistance to chlorine[b]	Relative infectivity[c]	Important animal source
Protozoa					
Acanthamoeba spp.	High	May multiply	Low	High	No
Cyclospora cayetanensis	High	Long	High	High	No
Entamoeba histolytica	High	Moderate	High	High	No
Giardia intestinalis	High	Moderate	High	High	Yes
Naegleria fowleri	High	May multiply	Low	Moderate	No
Toxoplasma gondii	High	Long	High	High	Yes

[a]Detection period for infective stage in water at 20°C: short = up to 1 week; moderate = 1 week to 1 month; long = over 1 month. [b]When the infective stage is freely suspended in water treated at conventional doses and contact times and pH between 7 and 8. Low means that 99% inactivation at 20°C generally in <1 min, moderate 1–30 min, and high >30 min. [c]From experiments with human volunteers, from epidemiological evidence and animal studies. High means infective doses can be 1–102 organisms or particles, moderate 102–104 and low >104. [d]Includes enteropathogenic, enterotoxigenic, and enteroinvasive. eMain route of infection is by skin contact, but can infect immunosuppressed or cancer patients orally. *Vibrio cholerae* may persist for long periods in association with copepods and other aquatic organisms. fIn warm water

The human enteric viruses cause a wide range of diseases and symptoms. Viral etiology is rarely identified, even though viruses are believed to cause the majority of water borne illnesses (Griffin et al., 2003). The numbers of pathogenic microorganisms presented in surface waters are generally few and difficult to identify and isolate. The methods to detect pathogens are relatively laborious, require specialized personnel, and are not well suited for monitoring purpose.

In cases of human enteric viruses in water; interest has focused on indicator organisms that are nonpathogenic, rapidly detected, easily enumerated, similar survival characteristics to those of the pathogens and able to associate with the presence of pathogenic microorganisms. Treatment processes and watershed management strategies designed on the basis of bacteriological criteria do not necessarily protect against virus infection because viruses are generally more persistent in water environment and are not removed well by treatment processes (Havelaar et al., 1993); therefore many current studies have been directed toward identifying more and specifying more specific indicators of viral contamination.

The bacteriophages (bacterial viruses) have been proposed as useful alternatives to viral indicator; as their morphology and survival characteristics closely resemble those of some of the important human virus groups. Three types of bacteriophages have been proposed as specific indicators of viral contamination: the somatic coliphages, the F-specific RNA phages, and bacteroides fragilis phages (Morinigo *et al*., 1998; Havelaar *et al*., 1985; Jofre *et al*., 1989). Somatic coliphages are bacteriophages which consist of a capsid containing single or double stranded

DNA as the genome. These are violent phages which attach to lipopolysaccharide or protein receptors in the bacterial cell wall. Natural host strains of somatic coliphages include *Escherichia coli* or other closely related bacterial species. F-specific RNA bacteriophages consist of a simple capsid of cubic symmetry of 21-30 nm in diameter and contain single stranded RNA as the genome. These are infectious for bacteria which possess the F- or sex plasmid originally detected in Escherichia coli K-12. Bacteroides fragilis phages are DNA virus, about 60 nm in diameter, infecting by attaching to bacterial cell walls.

Several researches have been published on the use of bacteriophages as viral indicators for the presence of human enteric viruses. For example, F-specific RNA bacteriophage concentrations are highly correlated with those of enteric viruses in a wide range of water environments and water treatment processes (Havelaar *et al*., 1993). Bacteriophage Qβ is widely used as a surrogate for pathogenic waterborne viruses in Japan (Kamiko and Ohgaki, 1989; Urase *et al*.,1996; Otaki *et al*.,1998). On the other hand, research groups in Europe and United State widely used F-specific RNA bacteriophage MS2 (diameter 0.025 μm,) as a surrogate for virus removal (Adham *et al*., 1998; Fiksdal and Leiknes, 2006; Hu *et al*., 2003; Jacangelo *et al*., 1995; Langlet *et al*., 2009; van Voorthuizen *et al*., 2001; Zhu et al., 2005a; Zhu *et al*., 2005b). MS2 and Qβ belong to the Leviviridae family, having an icosahedral capsid with a linear single-stranded RNA genome (Grabow, 2001).

The F-specific RNA bacteriophage is enterobacteriaceae viruses of the Leviviridae family that are physically and genomically analogous to human enteric

viral pathogens found in sewage. They are abundant in sewage and easy to enumerate using well-standardized ISO methods (ISO10705-1) (Anon, 1996), making them a good prospective indicator of viral contamination in the marine environment. They have been proposed as indicators of viral contamination in the environment and used to model the behavior of human pathogen enteric viruses. They are divided into two genuses, Levivirus and Allolevivirus, each containing two fully characterized species based upon their genome organization and the antigenic specificity of their capsid proteins. The F-specific RNA bacteriophage, bacteriophage Qβ has been used extensively as a surrogate virus for waterborne viruses because of their morphological and structural resemblance to human enteric viruses (Matsui et al., 2003; Matsushita *et al*., 2005; Otaki *et al*., 1998; Shirasaki *et al*., 2007; Urase *et al*., 1996). The bacteriophage Qβ genome comprises of a single-stranded RNA molecule encapsulated in an icosahedral protein shell that is ca. 0.023 μm in diameter without an envelope.

The bacteriophage Qβ is used as a surrogate for waterborne viral pathogens because of its morphological similarity to hepatitis A virus and poliovirus, which are important targets for removal during drinking-water treatment. The bacteriophage Qβ was propagated for 22-24 h at 37°C in Escherichia coli F+. The bacteriophage Qβ culture was centrifuged (3000 x g) and then filtrated through 0.45 μm pore size membrane filter. The filtrate was purified using a centrifugal filter device to prepare the bacteriophage Qβ stock solution. Bacteriophage concentration was determined using plaque forming unit (PFU) assay with the agar overlay method (Adam, 1959)

and the bacterial host E.coli F+. Average plaque counts of triplicate plates prepared from one sample indicated the bacteriophage Qβ.

Some studies using other model and native viruses have been reported. The use of model viruses was discussed by Grabow (2001) and Templeton *et al*. (2008). Moreover, the study of Langlet *et al*. (2008a) confirms the use of MS2 and Qβ as model viruses meeting the right criteria of electro kinetic and aggregation features with respect to pH and ionic strength conditions compared with other two bacteriophages, GA and SP. However, MS2 and Qβ differed significantly among themselves, Qβ was observed to possess higher negative charge and a higher degree of hydrophobicity compared with MS2. Thus, under the conditions of insignificant viral aggregations, Qβ was removed less than MS2 phage, filtering with negative and hydrophilic membranes (Langlet *et al*., 2009). As a result, bacteriophage Qβ was used as a surrogate for waterborne pathogen in this experiment. Several studies compared the behavior of MS2 and Qβ toward various physicochemical treatment processes were shown in Table 2.4. Moreover, the virus removal efficiencies by using MF were shown in Table 2.5. The membrane elements under discussion are hexagonal in 3 centimeter in diameter, 10 centimeters height and tubular channels. These are available with three different pore sizes (1.0 μm, 0.5 μm and 0.1 μm).

Table 2.4 Behavior of MS2 and Qβ towards treatment processes (Source: Antiny *et al*., 2012)

Treatment process	Behavior of MS2 and Qβ	Results	Reference
Adsorption to solid surfaces such as cellulose, river sediments, suspended solids, kaolin and carbon black	analogous	Similar adsorption isotherm and relative activity of phage adsorption (**α**).	Sakoda et al., 1997
Sandy aquifer treatment	analogous	Similar breakthrough characteristics, rate coefficients for capture and release in sorption	Dowd et al., 1998
MF	analogous	Similar rejection behavior in cross flow mode, at different flux and cross flow velocities.	Herath et al., 2000
MF and UF	diverse	Qβ rejection is always lower than MS2, when the viral aggregation is absent or insignificant	Langlet et al., 2008a
Coagulation–Ceramic MF system	diverse	Removal efficiencies were different at different dosage levels, higher for MS2 at low dosage levels and for Qβ at high dosage levels.	Matsushita et al., 2006

TABLE 2.4. Behavior of MS2 and Qβ towards treatment processes (Source: Antiny *et al*., 2012) (continuous)

Treatment process	Behavior of MS2 and Qβ	Results	Reference
	diverse	Removal of infectious MS2 and Qβ was similar. But the removal ratio of total MS2 was nearly 2-log higher than total Qβ removal, due to selective interaction with cake layer.	Shirasaki et al., 2009
Aluminum coagulation process	diverse	Extent of inactivation was higher for Qβ compared to MS2	Matsushita et al., 2004
	diverse	Removal ratio of infectious Qβ was approximately 2-log higher than infectious MS2. But the removal of total MS2 and Qβ was analogous.	Shirasaki et al., 2009
UV radiation	diverse	MS2 was more resistant to UV compared with Qβ	Hijnen et al., 2006; Simonet and Gantzer, 2006; Blatchley et al., 2008

Table 2.5 Removal efficiencies reported for viruses by MF (Source: Antiny *et al*., 2012)

Type	Feed water	Scale	Membrane speccification	Virus concentration in the feed	LRV	Reference
MS2	Surface water[a]	Pilot	MF 0.2 µm (PP)	10^{12} pfu/mL	2.0–3.0	Adham and
	Surface water[b]	Pilot	MF 0.2 µm (PP)	10^{12} pfu/mL	0–1.0	Jacangelo, 1994;
		Pilot.	MF 0.2 µm (PSD)	10^{12} pfu/mL	0–1.0	Jacangelo, Adham
	Surface waterc	Pilot	MF 0.2 µm (PP)	10^{12} pfu/mL	1.0	et al., 1995
		Pilot	MF 0.2 µm (PSD)	10^{12} pfu/mL	2.0–3.0	
MS2	Buffered DI	Bench	MF 0.2 µm (PS)	na	<1	Jacangelo et al., 2005
	water[d]	Bench	MF 0.1 µm (PVDF)	na	<1	
MS2	Buffered DI water[d]	Bench.	MF 0.1 µm (PVDF)	10^{3}–10^{6} pfu/mL	1.79 ±0.09	Langlet et al., 2009
Somatic	Wastewater	Pilot	MF 0.2 µm (PP)	$2.3x10^{5}$–$2.8 x10^{6}$	4.8-5.9	Farahbakhsh and
coliphages				cfu/mL		Smith, 2004
Poliovirus 1 (PV1)	Buffered DI	Bench	MF 0.1 µm (PVDF)	na	0–1.0	Jacangelo et al., 2005
	water[d]					
Qβ	Buffered DI	Bench	MF 0.1 µm (PVDF)	10^{3}–10^{6} pfu mL^{-1}	1.25±0.05	Langlet et al., 2009
	water[e]					

Note. na = not available.

[a]Sourced from Bull Run reservoir (Portland, high-quality surface water), turbidity <1 ntu, TOC 1.3 mg/L. [b]Sourced from Lake Elsman (San Jose), more mineralized, turbidity 3.4 ntu, TOC 2.6 mg/L. [C]Sourced from the Seine River (Vigneux, lower quality surface water), turbidity 15 ntu, TOC 3.0 mg/L. [d]Seeded with microorganisms, pH 7. [e]Seeded with microorganisms, pH 6.7(Source: *Antiny et al*., 2012

CHAPTER III

METHODOLOGY

3.1 Ping River

Ping River, the main river in north Thailand, is one of the headstreams of the Chao Phraya River. The river contains suspended solid about 40-80 NTU, draining 33,896 km^2 of land area. This river is 569 Km long, and has its source in the mountains near Chiang Dao, in the northernmost part of Chiang Mai Province. It flows southward through the city of Chiang Mai and provides the surrounding rural countryside with its much needed water for irrigation of rice paddies, gardens and crops.

Figure 3.1 Sampling point in Ping River, Chiang Mai Province

Figure 3.2 Sampling point in Ping River, Chiang Mai Province

3.2 Experimental Procedure

About 240 liters of Ping River were collected in December 2011, transported in polyethylene tanks and stored at 4°C before being analyzed within 24 hours. Physical-chemical parameters and DOM parameters of Ping River were analyzed.

The experimental procedures are shown in the following steps and conclusively described in the diagram in Figure 3.3 and these are described below. Raw water were analyzed for pH, temperature, turbidity, alkalinity, conductivity and DOM. Raw water were divided for 3 experiments.

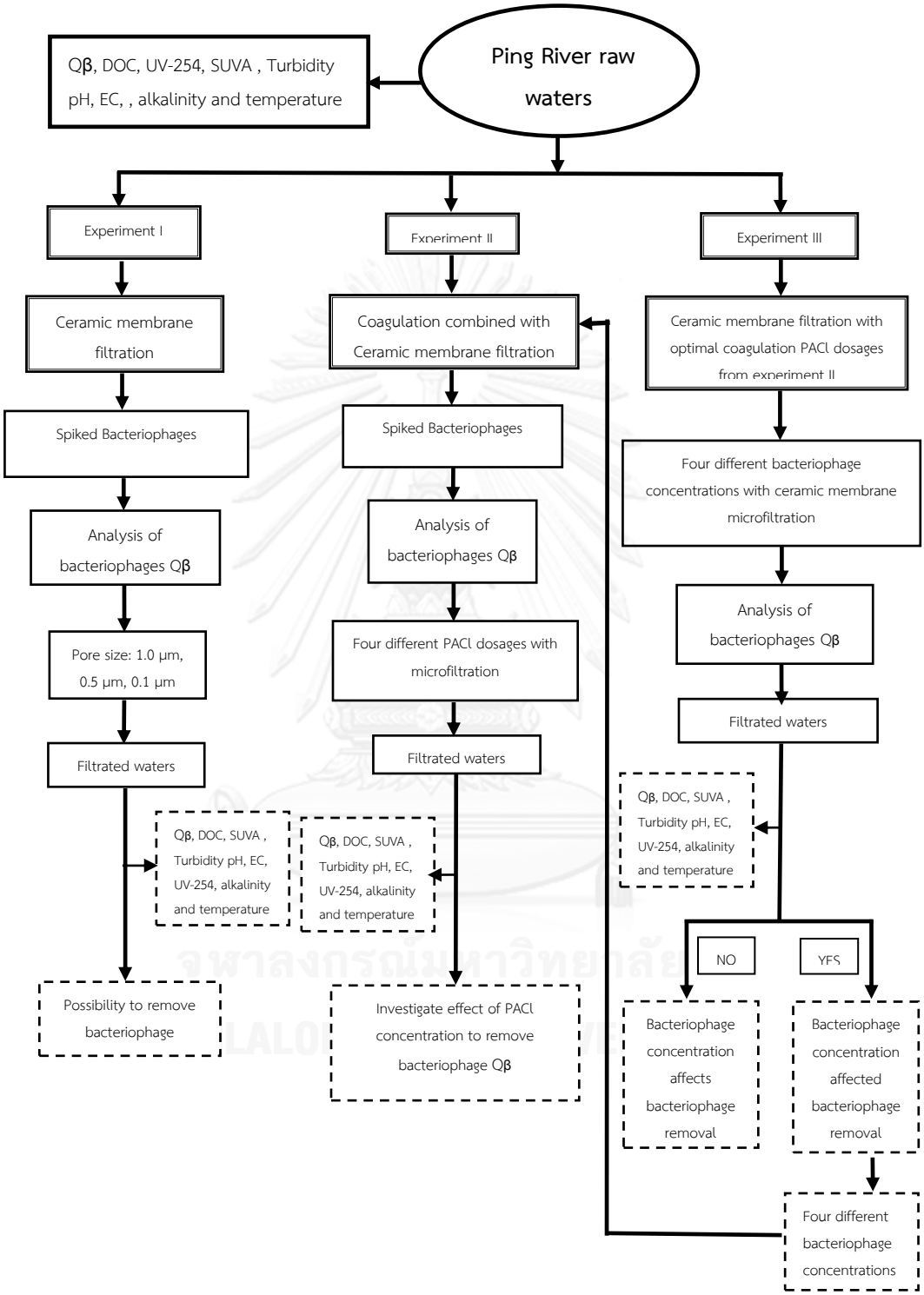


Figure 3.3 Diagram of overall experiment procedure for bacteriophage reduction.

For the first experiment, raw water was spiked bacteriophage Qβ (NBRC 20012) which obtained from NITE Biological Resource Center (NBRC, Chiba, Japan). Bacteriophages Qβ were analyzed in overlay plaque assay by using *Salmonella typhimurium* WG 49 as host strains.

The spiked-surface water was filtered through 1.0 μm, 0.5 μm, and 0.1 μm ceramic membrane filtration. Filtrated water were analyzed the bacteriophages Qβ concentration, the procedures adopted were the standardized protocol (ISO, 1997). Bacteriophage Qβ concentration reported in plaque forming unit (PFU). WG49 host strain was incubated in Tryptone-yeast extract-glucose-broth for 18±2 h at 37 °C with shaking at 150 rpm. About 1 milliliter of dilution sample and 1 milliliter of exponentially growing WG 49 host culture were added to molten Semi-solid tryptone-yeast extract-glucose agar. Mixed and poured in a petri dish. The overlays were incubated over night at 37 °C. When higher bacterial background flora may interfere with growth of the host and replication of phages, the addition of nalidixic acid and kanamycin is recommended to suppress contaminant growth**.** Bacteriophage Qβ concentration reported in plaque forming unit (PFU). The bacteriophage Qβ concentration at 1 (PFU/mL) was a value of detection limit. In addition, DOC, UV-254, and SUVA were measured for investigate the DOM surrogate reduction. The results in this part represent the reduction efficiency of bacteriophage Qβ by ceramic membrane microfiltration.

In the second part, spiked-surface water was used to determine the most achievable PACl dosage for reduce bacteriophage Qβ in raw water when the PACl coagulation was combined with ceramic membrane filtration. Polyaluminium Chloride (PACl) was used as a coagulant in coagulation and varied to 1.5, 2.0, 2.5, 3.0 mg-Al/L. The same experiment as for filtrated waters in the first experiment was conducted. The results in the second experiment represent the efficiency of bacteriophage Qβ reduction by the ceramic membrane microfiltration combined with coagulation.

In the third part, the different initial bacteriophage Qβ were used to investigate the efficiency of bacteriophage Qβ reduction by the ceramic membrane microfiltration combined with the most achievable PACl dosage from the second part. The bacteriophage Qβ concentrations were $5.0x10^{5}$, $4\ x10^{6}$, and $8.0\ x10^{7}$ PFU/mL). The same experiment as for filtrated waters in the previous part was conducted. The results in this experiment represent the effect of bacteriophage Qβ concentration in feed water for the reduction efficiency of ceramic membrane filtration combined with coagulation.

The obtained results from all experiments could be used to represent the reduction bacteriophage Qβ efficiency using ceramic membrane microfiltration combined with coagulation.

3.3 The coagulation combined with ceramic membrane microfiltration

3.3.1 Ceramic membrane module preparation

Three different pore sizes of ceramic membrane; 1.0 μm, 0.5 μm and 0.1 μm provided by Metawater Co., Ltd., Japan were used. The dimension of each ceramic membrane module is 3 centimeters in diameter, 10 centimeters height and 55 tubular channels, as shown in Figure 3.4.

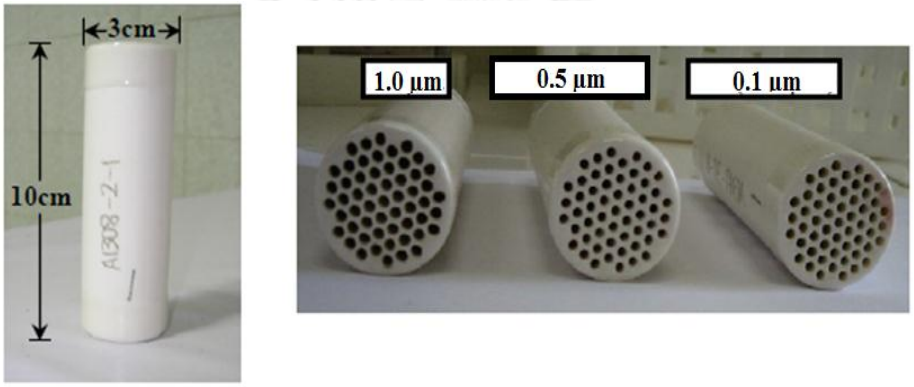


Figure 3.4 Ceramic membrane modules

All membrane modules were prepared by the following method as describe by Katayama *et al*., 2002. The membrane modules were boiled for 10 minutes before being used in filtration tests. After processing, in order to remove the organic and inorganic fouling from the membrane surfaces, the cleaning procedure was performed by submerging ceramic membrane module into the acid and base solutions in the following order: 1% nitric acid solution and 0.3%(as available chlorine) sodium hypochlorite solution, each for one hour.

3.3.2 The operation of ceramic membrane microfiltration with coagulation.

Some of the spiked-surface water samples were filtrate through ceramic membrane microfiltration unit. The schematic figure of the ceramic membrane microfiltration process was shown in Figure 3.5. Ceramic membrane modules were installed in a stainless steel vessel vertically, and operated in the dead end mode. Raw water was spiked with Qβ and mixed by jar test apparatus at mixing speed 150 rpm for 60 second. About 3 liters of Spiked-surface water was poured to pressurized tank. PACl dosage of 1.5, 2.0, 2.5 and 3.0 mg-Al/L were added to spiked-Qβ raw water and immediately mixed. The purged pressure was controlled by the adjustment from the pressure regulator of nitrogen gas. The spiked-surface water was pressurized by nitrogen gas to the membrane housing and filtrated from inside to outside through ceramic membrane. After that, the coagulated water in a pressurized tank was allowed to 8-meters-nylon tube prior, flowing to the bottom end of the ceramic membrane module that feeding control was regulated by nitrogen gas controlled pressure at 0.2 MPa as shown in Figure 3.5.

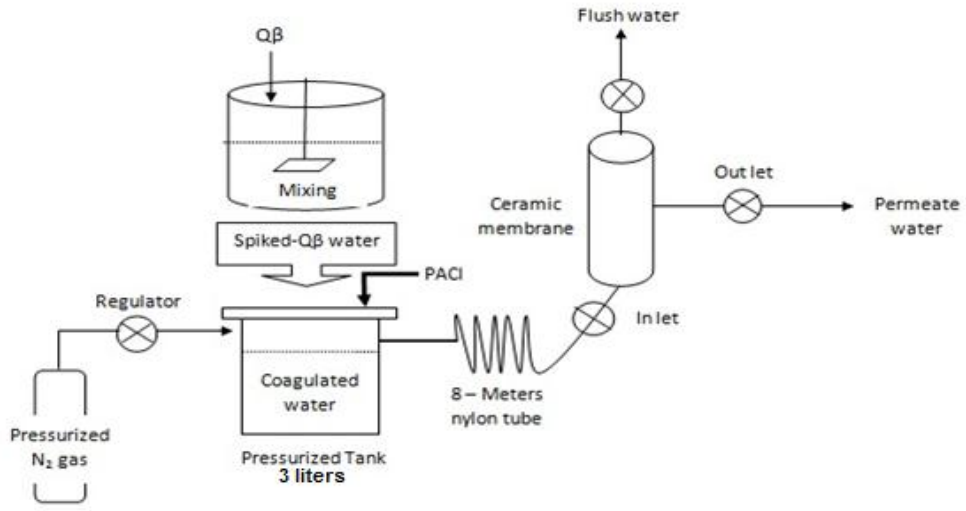


Figure 3.5 Schematic diagram of coagulation combined with ceramic membrane microfiltration.

Spiked-Qβ surface water and permeate water were collected for evaluate Qβ reduction efficiency. After processing, in order to remove the organic and inorganic fouling from the membrane surfaces, the cleaning procedure as describe by Katayama et al., 2002 was performed to prepare the membrane modules for the next experiment.

3.3.3 Flux measurement

Initial flux was measured by measuring the filtration time of 2 liter of filtrate from RO water (average value from 2-3 times) by the controlled pressure at 0.2 MPa. The initial flux measurement was performed before sample filtration of every batch experiment. Similarly, water sample flux was measured as the same procedure as the initial flux measurement.

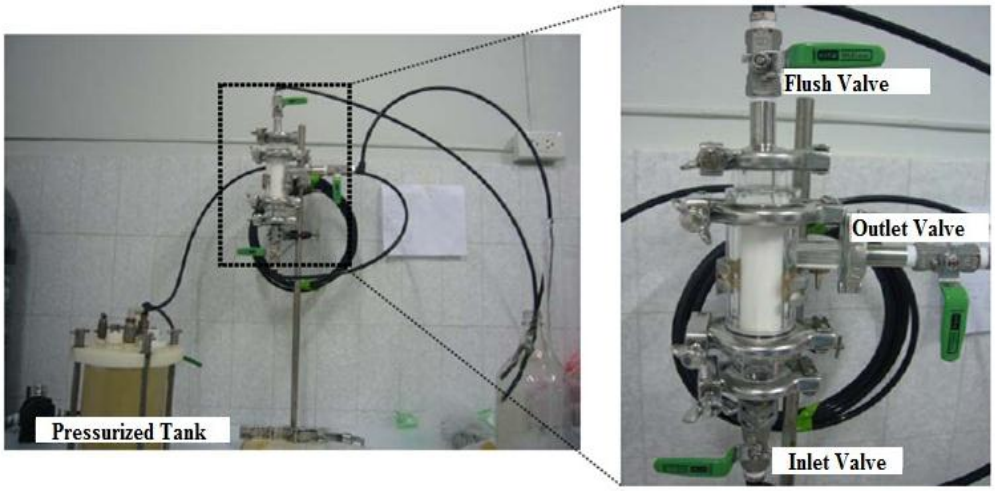


Figure 3.6 The experimental set-up of ceramic membrane microfiltration with coagulation.

3.4 Analytical Methods

3.4.1 Physico-chemical parameters

The water samples were analyzed for pH, turbidity and alkalinity, UV-254, DOC and SUVA. The summary of analytical methods and standards used for analyzing the mentioned parameters demonstrated in Table 3.1. These parameters are described below. The analyzed parameters were done by duplicate samples. The results of these analyses should be within ±5%, or corrective action is necessary.

3.4.1.1 pH

pH was directly measured by a Model F-21 Horibra pH-meter with an accuracy of ± 0.01 pH unit. The unit was daily calibrated with buffer solutions at pH 4.00, 7.00 and 9.00.

3.4.1.2 Temperature

Temperature was directly measured by Horiba Thermometer, Model D-13E.

3.4.1.3 Turbidity

Turbidity was measured by the HACH Turbidity meter Model 2100.

3.4.1.4 Alkalinity

Alkalinity was measured in accordance with Standard Method 2320 B.

3.4.1.5 Electro Conductivity

Electro conductivity was directly measured by WTW Conductivity meter, Model cond.330i

3.4.2 DOM surrogate parameters

3.4.2.1 DOC

DOC of water samples were measured in accordance with standard method 5310 Total Organic Carbon (TOC); section 5310 C Persulfate-Ultraviolet Oxidation Method by using O.I. analytical 1010 TOC Analyzer. Water samples were filtered through a 0.45 μm filter prior to measurement. Milli-Q water (ELGA) was used on every sample for clean system and blank sample preparation. The analysis of DOC was conducted with two replications for each sample.

3.4.2.2 UV-254 nm

UV-254 of water samples were measured in accordance with standard method 5910 B Ultraviolet Absorption Method. The samples were measured by Perkin-Elmer Model Lambda 25, UV/VIS spectrophotometer.

3.4.2.3 Specific ultraviolet absorption (SUVA)

SUVA of water samples was calculated from the ratio of UV absorbance at 254 nm to DOC value in mg/L.

Table 3.1 The summary of analytical methods and instruments

Parameters	Analytical methods	Standards	Analytical Instruments
Temperature	Direct measurement	-	Horiba Thermometer, ModelD-13E
pH	Direct measurement	-	Horiba pH meter, Model F-21
EC	Direct measurement	-	WTW Cond. meter, Modelcond.330i
Turbidity	Direct measurement	-	HACH, 2100 Turbidity Meter

Parameters	Analytical methods	Standards	Analytical Instruments
Alkalinity	Titration Method	Standard method 2320B*	-
UV-254	Ultraviolet Absorption Method	Standard method 5910 B*	Jasco, Model UV-530, UVspectrometer
DOC	Wet Oxidation Method	Standard method 5310C*	O.I. analytical 1010 TOC Analyzer

3.5 Analysis of Microorganisms

To evaluate the virus reduction performance of ceramic membrane microfiltration at different pore sizes, the F-specific RNA bacteriophage (Qβ) have been used as possible indicator for enteric viruses as their morphology and survival characteristics closely resemble to some of the important human virus groups. The reduction efficiency of ceramic membrane microfiltration is usually reported as a Log removal value, LRV (Bennett, 2008):

$$LRV = \log C_i / C_p$$

Where Ci is the bacteriophage Qβ initial concentration and Cp is the concentration of bacteriophage Qβ in filtrate. Regulations and guidelines for drinking

water and water recycling specify a target LRV that reduces the risk associated with exposure to the pathogen to a tolerable level. For example, the specified inactivation or removal efficiencies for various pathogens defined in the USEPA Enhanced Surface Water Treatment Rule (ESWTR) is 2 LRV (i.e., 99% removal) for *Cryptosporidium parvum*, 3 LRV (i.e., 99.9% removal) for *Giardia Lamblia*, and 4 LRV (i.e., 99.99% removal) for viruses (Bennett, 2005).

The bacteriophage Qβ was spiked in all experiments by according to the recommendation of U.S. Environmental Protection Agency, which regarded the control of the quality of treated surface water by membrane filtration. Virus feed concentration has to be sufficiently high to allow the demonstration of up to 6.5 log removal when the surrogate is removed to the detection limit. To achieve up to 6.5 log removal, $8x10^6$ PFU/mL was the initial bacteriophage Qβ concentration in this experiment.

3.5.1 Analysis total coliform and *Escherichia coli* (*E. coli*)

Analysis of total coliform and *Escherichia coli* (*E. coli*) Total coliform and *E. coli* were analyzed by single agar layer method using Chromocult Coliform agar (Merck, USA). Samples were dilution in LB broth if necessary and add 1 ml of the water sample or diluted sample by LB Broth in the petri dish. Pour approximately 15 ml of the agar solution into petri dish. The microbes were assayed after incubated at 37 °C for 18-24 hrs. This agar performed three different colored colonies. Salmon to

red colonies and dark-blue to violet colonies were counted as total coliforms. Dark-blue to violet colonies were counted as *E.coli*. The concentration of microbes was reported as CFU/ml (Colonies Forming Unit/ ml).

3.5.2. Analysis of Bacteriophages

F-specific bacteriophage Qβ

Bacteriophage Qβ (NBRC 20012) obtained from NITE Biological Resource Center (NBRC, Chiba, Japan) as a model virus. The bacteriophage Qβ genome comprises a single-stranded RNA molecule encapsulated in an icosahedral protein shell that is ca. 0.023 μm in diameter without an envelope. Bacteriophage Qβ is used as a surrogate for waterborne viral pathogens because of its morphological similarity to hepatitis A virus and poliovirus, which are important targets of removal during drinking-water treatment.

Bacteriophage Qβ was conducted by following the protocol by Katayama *et al*. (2002). Briefly, Indigenous F-specific RNA bacteriophages Qβ were analyzed in overlay plaque assay by using *Salmonella typhimurium* WG 49 as host strains. WG49 host strain was incubated in Tryptone-yeast extract-glucose-broth for 18±2 h at 37 °C with shaking at 150 rpm. About 1 milliliter of dilution sample and 1 milliliter of exponentially growing WG 49 host culture were added to molten Semi-solid tryptone-yeast extract-glucose agar. This was mixed and poured in a petri dish. The higher bacterial background flora may interfere with growth of the host and

replication of phages, the addition of nalidixic acid and kanamycin is recommended to suppress contaminant growth. The overlays were incubated over night at 37 °C. Qβ concentration reported in plaque forming unit (PFU).

Research involving viral systems necessitates precise quantification. Currently, the standard quantitative methodology for phage preparations is the traditional plaque assay (Sambrook *et al*., 1989). However, significant limitations of this method include: (i) , the requirement for extensive hands-on time (≥5 h) for completion of the assay; (ii) a limited dynamic range of one log (30–300 plaques/plate). Average plaque counts from triple plates prepares from one sample indicated the virus concentration that was illustrated standard deviation less than 30%. Detection limit was 1 PFU/mL.

3.5.3. Host Preparation

WG 49: *Salmonella typhimurium*

Working culture

Stock from freezer was diluted around 10^{-7} – 10^{-8}. Working culture was prepared by using pour plate technique using MacConkey agar as a medium. The culture plate was incubated overnight at 37 °C. The working culture plate was kept in the refrigerator at 4 °C for 3 weeks. The concentration of red colonies in MacConkey

agar must be more than 10^8 CFU/mL and the concentration of white colonies in the agar should be less than 10% of the colonies in agar.

Inoculums culture

3-4 red colonies with agar were picked up from working culture plate and added into the 10 ml TYGB. The culture was incubated with shaking for 24 hours at 37 °C. 10 ml of inoculum culture was used with 500 ml of TYGA agar and used for preparation of working plate for next step.

K12: *E. coli* K12 A/λ (F+)

Working culture

Stock of *E. coli* K12 A/λ (F+) from the freezer that was previously thawed at room temperature was added in 10 ml of LB broth. The culture was incubated at 37 °C with shaking at 150 rpm for 2-3 hours. 100 µl of culture was spread on the solidified bottom agar (LB agar 2) that was prepared on the same day of preparation of working culture. The culture plate was incubated overnight at 37 °C. The working culture plate was kept in the refrigerator at 4 °C for 3 weeks.

Inoculum culture

Colonies from the working plate were smeared and added into 10 ml of LB broth. The culture was incubated at 37 °C while shaking for 2-3 hours. 10 ml of inoculum culture was used with 500 ml of top agar (LB agar 1) and was used for preparation of working plate for next step.

3.5.4 Culture Media, Reagents and Diluents

3.5.4.1 Modified Scholten's broth (MSB), Modified Scholten's Agar (MSA) and Semi-solid modified Scholten's agar (ssMSA)

MSB : Broth for inoculum culture

MSA : Bottom Agar Media

ssMSA : Upper Agar Media

The ingredients were shown in Table 3.2. The ingredients were dissolved in hot water. The mediums were distributed into bottles or vials and sterilized in the autoclave at 121 ℃ for 15 min.

Table 3.2 The ingredients of MSB, MSA, and ssMSA

Components	Unit	MSB (Broth)	ssMSA (Upper layer agar)	MSA (Bottom layer agar)
Peptone	g	10	10	10
Yeast extract	g	3	3	3
Meat extract	g	12	12	12
NaCl	g	3	3	3

Components	Unit	MSB (Broth)	ssMSA (Upper layer agar)	MSA (Bottom layer agar)
Na_2CO_3 solution (150 g/l)	Ml	5	5	5
MgCl2 solution (100 g/ 50 ml)	Ml	0.3	0.3	0.3
Bacto agar	g	-	7	15
CaCl2* 14.7 g/ 100 ml	ml	-	6	6

Note: * $CaCl_2$ solution was pre-warmed and added to top agar prior adding agar to petri dish.

3.5.4.2 LB Broth base for dilution

LB Broth (Invitrogen)	20 g
Milli-Q water	1000 ml

The LB Broth was dissolved in the Milli-Q water while heating gently. The media was transferred to the vials and autoclaved at 121 °C for 15 min. The solution was stored for experiment.

3.5.4.3 Na_2CO_3 solution

The 15 g of Na_2CO_3 was dissolved in 100 ml of Milli-Q water. The solution was decontaminated by 0.22 µm membrane filtration.

3.5.4.4 $MgCl_2$ solution

The 100 g of $MgCl_2.6H_2O$ was dissolved in 50 ml of Milli-Q water. The final concentration of Mg^{2+} in this solution was 4.14 mol/L. The solution was sterilized by autoclaving and stored at room temperature in the dark.

3.5.4.5 $CaCl_2$ solution

The 1 M of $CaCl_2$ was prepared as stock solution by dissolving 14.7 g of $CaCl_2.2H_2O$ in 100 ml of Milli-Q water by gentle heating. The solution was decontaminated by 0.22 µm membrane filtration and stored at 4±2 °C: for maximum of 6 months.

3.5.4.6 Trytone-Yeast extract-Glucose-Broth (TYGB), Trytone-Yeast extract Glucose-Agar (TYGA), and Semi-solid Trytone -Yeast extract-Glucose-Agar (ssTYGA)

TYGB : Broth for inoculum culture

TYGA : Bottom agar media

ssTYGA : Upper agar media

The ingredients were shown in Table 3.3. The ingredients were dissolved in hot water. The mediums were distributed into bottles or vials and sterilized in the autoclave at 121 ℃ for 15 min.

Table 3.3 The ingredients of TYGB, TYGA, and ssTYGA

Components	Unit	TYGB	ssTYGA (Upper layer agar)	TYGA (Bottom layer agar)
Tryptone	g	10	10	10
Yeast extract	g	1	1	1
Glucose	g	1	1	1
NaCl	g	8	8	8
$CaCl_2$ 0.3 mg/L	ml	1	1	1
$MgSO_4$ 0.15 mg/L	ml	1	1	1
Bacto agar	g	-	9	15

CHAPTER IV

RESULTS AND DISCUSSION

4.1 Characteristics of raw surface water

The raw surface water was collected from Ping River. The physical characteristics including pH, temperature, conductivity, turbidity and alkalinity were analyzed. Dissolved organic matter (DOM) surrogate parameters (DOC, UV-254, and SUVA) were also investigated. The summary of the characteristics of raw surface waters from Ping River is presented in Table 4.1.

Table 4.1 Characteristics of raw surface waters from Ping River

Parameters	Raw surface waters (n=3)
pH	7.69±0.02
Temperature (C^{o})	24.7±0.25
Conductivity (μS/cm)	222.0±4.72
Turbidity (NTU)	41.77±4.4
Alkalinity (mg/L as $CaCO_3$)	108.9±1.8
DOC (mg/L)	2.324±0.03
UV-254 (cm^{-1})	0.086±0.001
SUVA (L/mg-m)	3.58±0.09

Parameters	Raw surface waters (n=3)
Qβ concentration (PFU/mL)	ND
***E. coli* (CFU/mL)**	27±0.05
Total Coliform (CFU/mL)	296±1.52

*ND : not detected

The average pH and alkalinity values of Ping River water were 7.69 and 108.87 mg/l $CaCO_3$, respectively. It can be noticed that pH of raw water sources was nearly neutral. In order to prevent pH drop during coagulation/flocculation process, the conventional water coagulation that use alum (aluminum sulfate) as coagulant was generally required the additional alkalinity in case of low alkalinity raw water. Alkalinity is 108.9 enough value. PACl are synthetic polymers dissolved in water and reacted to form insoluble aluminium poly-hydroxides. Solutions of PACl are not as acidic as alum. Therefore, PACl was induced as coagulant without pH adjustment in this study.

The DOC and UV-254 were 2.324 mg/L and 0.086 cm^{-1}, respectively. The value of DOC in water used to indicate the aromatic and aliphatic hydrocarbons. SUVA was used as an index of humic in water, calculated from the ratio of UV-254 absorbance and DOC values. SUVA values of less than 3 L/mg-m signify water containing mostly non-humic material indicated the presence of organic matter of lower average molecular weight (AMW) with more fulvic character. SUVA values of 4-5 L/mg-m are typical of waters containing primarily humic material (Edzwald and Van

Benschoten, 1985). As the SUVA values demonstrated in table 4.1, the average SUVA value observed was 3.58 L/mg-m. It can be stated that Ping River water mostly contains humic material. Similarly to the report of Leenheer *et al*. (2001) stated that the DOM in surface water is mainly composed of humic substances (50%--65%) and possible to reduce by coagulation process. The turbidity of Ping River water was 41.77 NTU. From Provincial Waterworks Authority, Thailand (PWA, 2013) stated that the standard of turbidity of water supply was at 5 NTU. It can be indicated that this water cannot consume directly for potable water. It is required to treat for turbidity reduction before using as potable water. The amount of solid contained in Ping River water may have affected the aggregates (flocs) formation in the coagulation/ flocculation process that was proposed in the next topic.

Microbial indicators, Total Coliform Bacteria and *E.coli* were used as indicators for determining the fecal pollution reduction. Total coliform and *E.coli* was found from Ping River water in amount of 296±1.52 CFU/mL and 27±0.05 CFU/mL, respectively. From the results, it could be concluded that the microbial quality of the water sources was useless and unacceptable for human consumption due to fecal pollution. The standard of microbial indicators of water supply was set faecal coliforms at 0 CFU/100ml by Province Waterworks Authority, Thailand (PWA, 2013).

The concentration of bacteriophages Qβ in raw surface water samples were observed under the detection limit. The detection limit of the plaque assay which corresponds to the smallest amount of phages that could be detected was 1 PFU/mL. As results, it is necessary to apply more water sample volume adding for

plaque assay and/or apply virus concentration method in order to increase bacteriophages concentration in water samples prior plaque assay. Thus, bacteriophage Qβ was spiked in all experiments by according to the recommendation of U.S. Environmental Protection Agency, which regarded the control of the quality of treated surface water by membrane filtration. Virus feed concentration has to be sufficiently high to allow the demonstration of up to 6.5 log removal when the surrogate is removed to the detection limit.

4.2 Reduction of bacteriophage Qβ concentration by ceramic membrane microfiltration

The reduction of bacteriophage was investigated by considered the reduction of bacteriophage concentration in filtrated water. In addition, the reduction of DOM surrogate parameters including DOC, UV-254, and SUVA also were investigated and discussed.

To evaluate the bacteriophage removal performance of ceramic membrane microfiltration at different pore sizes (0.1 μm, 0.5 μm and 1.0 μm), the F-specific RNA bacteriophage Qβ have been used as possible indicator for enteric viruses as their morphology and survival characteristics closely resemble to some of the important human virus groups. The performance of bacteriophage Qβ reduction of spiked-surface water, using three different membrane pore sizes as measured by overlay plaque assay method. The removal efficiency of ceramic membrane microfiltration is

usually reported as a Log removal value, LRV = log Ci /Cf (Bennett, 2008). Where Ci is the bacteriophage Qβ initial concentration and Cf is the concentration of bacteriophage Qβ in filtrate. Regulations and guidelines for drinking water and water recycling specify a target LRV that reduces the risk associated with exposure to the pathogen to a tolerable level.

In this study, the turbidity of spiked-Qβ raw water was 41.77 NTU. The DOC and UV-254 were 2.20 mg/L and 0.086 cm-1. Bacteriophage Qβ was spiked to raw surface water at 8.0x106 PFU/mL and mixed by jar test apparatus at mixing speed 150 rpm for 60 second. Then, spiked-Qβ raw surface water was poured to pressurized tank and fed into the MF module in dead-end mode. The fed control was regulated by nitrogen gas controlled pressure at 0.2 MPa. Bacteriophage Qβ was analyzed by overlay plaque assay using *Salmonella typhimurium* WG 49 as host strain. The removal efficiencies obtained for bacteriophage Qβ at the different membranes pore sizes (1.0 μm, 0.5 μm and 0.01 μm) are presented in Table 4.2

Table 4.2 shows the effect of pore size of ceramic membrane microfiltration on bacteriophage Qβ removal, bacteriophage Qβ concentration were reduced from $8x10^6$ PFU/ml in spiked surface water to $1.78x10^6$ PFU/ml, $1.18x10^6$ PFU/ml and $9.33x10^5$ PFU/ml by ceramic membrane microfiltration at pore-size of 1.0 μm, 0.5 μm and 0.1μm, respectively. The larger pores could be an explanation of the very poor virus reduction, since the diameter of bacteriophage Qβ (approximately 0.023μm) was smaller than the pore size of the ceramic membrane. It illustrated that the pore size

of ceramic membrane could be insignificant for removing bacteriophage Qβ in raw surface water.

Table 4.2 Removal efficiency of bacteria and Qβ by ceramic membrane microfiltration at different pore sizes.

samples	Bacteria (CFU/mL)				Bacteriophage (PFU/mL)	
	E. coli	Log removal	Total coliform	Log removal	Bacteriophage Qβ concentration	Log removal
Raw water	27	-	2.96×10^{2}	-	8.00×10^{6}	-
1.0 μm	4.0	0.8	1.50×10^{1}	1.3	1.78×10^{6}	0.7
0.5 μm	ND*	1.4	ND	2.5	1.18×10^{6}	0.8
0.1 μm	ND	1.4	ND	2.5	9.33×10^{5}	0.9

*ND : not detected

The highest bacteriophage Qβ reduction efficiency of 0.9 log removal was achieved for the 0.1 μm ceramic membrane pore size. These results are related to the studied of Matsushita *et al*. (2005) which reported that low reduction in virus levels was observed in an experiment of membrane filtration run without PACl. The low bacteriophage Qβ log removal was presented at all pore sizes due to some of bacteriophage Qβ were rejected by aggregate with small suspended matter in water. The reduction of bacteriophage by a membrane pore depends on bacteriophage size to pore diameter. Membrane pores larger than the bacteriophage size can also refuse bacteriophage due to the adsorption or electrostatic repulsion, which occurred from a negative charge on the membrane surface or within its pores and a negative charge

on a bacteriophage (Ahmed El-Hadidy, 2011). The results were demonstrated the low reduction efficiency when bacteriophage Qβ retention on and/ or in membrane surface was relied on only sieving mechanism (Antony *et. al.,* 2012).

Antony *et. al* (2012) proposed that membrane filtration cannot be expected to be as effective a barrier for virus-sized particles based on the nominal pore size, some virus removal was evident. Therefore, several studies suggested that virus rejection by membrane is improved by the use of coagulants. Efficient virus rejection was reported in a combined process of pre-coagulation or flocculation with membrane filtration (Fiksdal and Leiknes, 2006; Matsui *et al*., 2003; Matsushita *et al*., 2005; Shirasaki *et al*., 2009a; Shirasaki *et al*., 2009b; Zhu *et a*l., 2005a)

Figure 4.1 shows the effect of ceramic membrane pore sizes on microbial indicators removal. Total and fecal coliforms have been used extensively for many years as indicators for determining the sanitary quality of natural water. The concentration of microbes was reported as Colonies Forming Unit/ ml (CFU/ml). The detection limit in this experiment was 1.0 CFU/ml. Total coliform and *E. coli* were found from surface water in amount of 296±1.52 CFU/ml and 27±0.05 CFU/ml, respectively. From the results obtained from this section, ceramic membrane microfiltration at 1.0μm pore size could fairly remove total coliform and *E. coli* while the smaller pore sizes ceramic membrane could remove total coliform and *E.coli* completely.

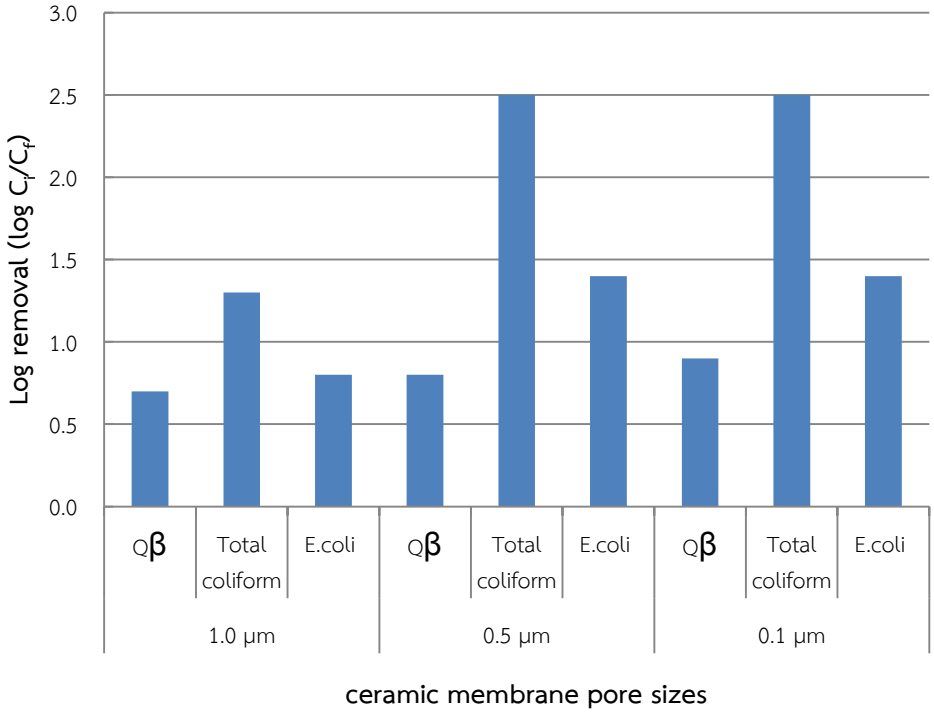


Figure 4.1 Effect of ceramic membrane pore sizes on microbial indicators removal.

According to the results, ceramic membrane microfiltration could be efficiently used for fecal pollution treatment, because coliform bacteria are larger than the absolute pore size of the membranes (0.6–1.2 µm in diameter by 2–3 µm in length). In the parts of microbial quality of the water, it could be concluded that the water quality was poor and unacceptable for human consumption at 1.0µm pore size due to faecal pollution. Drinking-water supplies should be protected from contamination. The standard of microbial indicators of water supply was set the maximum limit for no risk of faecal coliforms at 0 CFU/100ml by Province Waterworks Authority, Thailand (PWA, 2013).

4.3 Reduction of bacteriophage Qβ concentration by ceramic membrane microfiltration with PACl coagulation.

Since the diameter of the bacteriophage Qβ (0.023 μm) was smaller than the ceramic membrane pore sizes (Matsui *et al*., 2004), the lower bacteriophage Qβ removal than 1 log removal was observed in previous study. According to several studies, although ceramic membrane cannot be expected to be as effective a barrier for virus sized particles based on the nominal pore size. The two mechanisms are recognized: (a) removal by the filtration effect of the fouling layer on the membrane surface, and (b) aggregation of virus particles into larger particles and thus improved rejection (Alice *et.al.,* 2012). In this section, coagulation was applied. Polyaluminium Chloride (PACl) was used as coagulant for coagulation combined with ceramic membrane microfiltration at different pore sizes (1.0 μm, 0.5 μm and 0.1 μm).

Since the reduction of bacteriophage Qβ concentration by ceramic membrane microfiltration provided low log removal, the PACl coagulation was applied with ceramic membrane microfiltration. As the detection limit was 1 PFU/ml, the ceramic membrane microfiltration with PACl coagulation was able to reduce bacteriophage Qβ concentration at all PACl dosage. The result show that the reduction of bacteriophage Qβ concentration was increased from 0.7 log removal by 1.0 μm ceramic membrane microfiltration to 3.8, 4.5, 5.8 and 6.9 log removal at PACl dosage 1.5, 2.0, 2.5 and 3.0 mg-Al/L, respectively. Similarly, the reduction of bacteriophage Qβ concentration by 0.5 μm ceramic membrane microfiltration also increased from 0.8 to 4.8, 5.7, 6.9 and 6.9 log removal at PACl dosage 1.5, 2.0, 2.5 and 3.0 mg-Al/L,

respectively. The smallest pore sizes (0.1 μm). presented the high log removal The log removal increased from 0.9 to 5.4, 6.9, 6.9 and 6.9 log removal at PACl dosage 1.5, 2.0, 2.5 and 3.0 mg-Al/L, respectively. The high bacteriophage Qβ log removal was obtained even at low PACl dosage. As shown in Table 4.3. The combination of ceramic membrane microfiltration with coagulation can developed the reduction of bacteriophage Qβ concentration in raw surface water. It can reduce bacteriophage Qβ concentration in raw surface water by increased up 3 log removal at all pore sizes and also increased with PACl dosage increasing.

Table 4.3 Reduction efficiency of bacteriophage Qβ by ceramic membrane microfiltration with coagulation at different pore sizes.

PACl doses	Bacteriophage Qβ (PFU/mL)			Log removal		
(mg-Al/L)	1.0 μm	0.5 μm	0.1 μm	1.0 μm	0.5 μm	0.1 μm
0.0	1.78×10^{6}	1.18×10^{6}	9.33×10^{5}	0.7	0.8	0.9
1.5	1.23×10^{3}	1.16×10^{2}	3.20×10^{1}	3.8	4.8	5.4
2.0	2.56×10^{2}	1.60×10^{1}	1.00×10^{0}	4.5	5.7	6.9
2.5	1.20×10^{1}	1.00×10^{0}	1.00×10^{0}	5.8	6.9	6.9
3.0	1.00×10^{0}	1.00×10^{0}	1.00×10^{0}	6.9	6.9	6.9

*Limit detection 1 PFU/mL

The effects of coagulation dosage on bacteriophage Qβ removal when the system operated with different pore sizes were shown in Figure 4.2. The ceramic membrane microfiltration with the lowest PACl dose (1.5 mg-Al/L) could not reduce

bacteriophage Qβ completely. Bacteriophage Qβ weakly absorbed to aggregates and pass through membrane. The ceramic membrane microfiltration with pore size of 0.1 μm achieved a log removal of 6.9 with PACl dosing at 2.0 mg-Al/L. Whereas, ceramic membrane microfiltration with pore size of 0.5 and 1.0 μm showed lower performances (5.7 log and 4.5 log, respectively). These results related to Matsushita *et al.* (2004) which reported that coagulant dosage strongly affected virus removal with the coagulation-MF hybrid system: the larger the coagulant dose, the greater the proportion of virus removed.

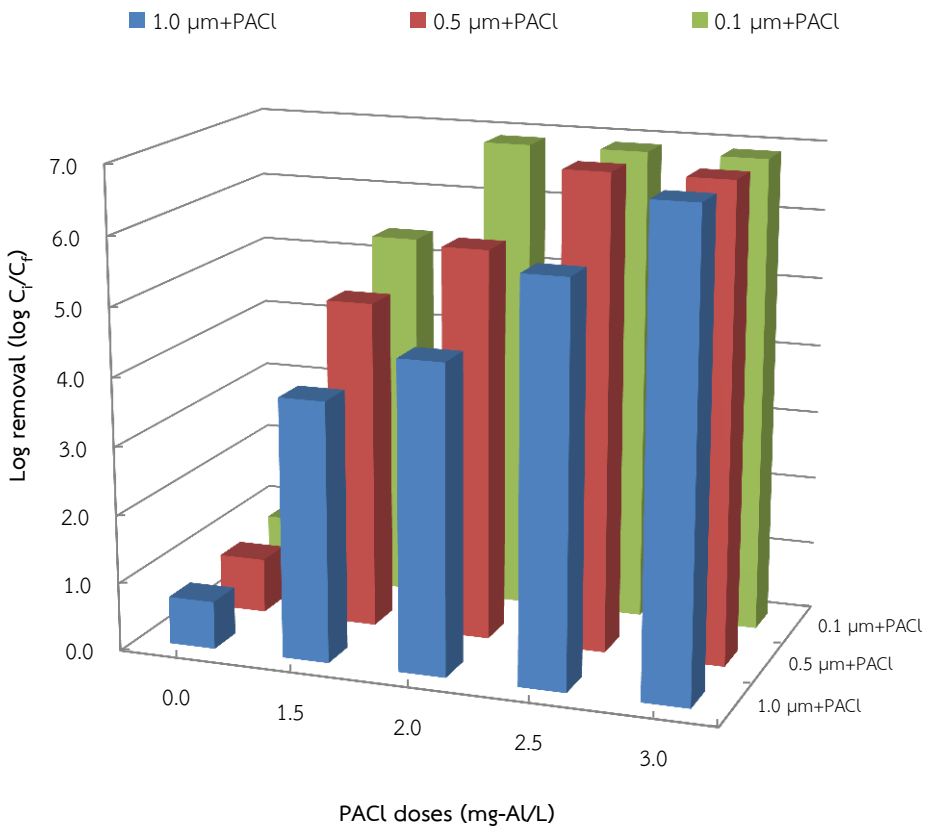


Figure 4.2 Reduction of bacteriophage Qβ concentration by ceramic membrane microfiltration with coagulation

From the review of literature, removal mechanisms of enteric viruses by membrane filtration will include size exclusion, electrostatic repulsion between charged membrane and charge virus and adsorption of viruses to the membrane material (Zeman and Zydney 1996). It is can be described that PACl coagulation with the ceramic membrane might contribute to the bacteriophage Qβ removal by adsorption or attraction on suspended PACl with enough adsorption potential, bacteriophage Qβ capture in the PACl cake layer and bacteriophage Qβ clogging in the constricted membrane pore including membrane physical sieving. In addition, clay particle naturally contained negative charge, when amino acid-RNA composition of bacteriophage Qβ is also negative charge at the pH of surface water (pH 7-8) (van Voorthuizen *et al.*2001). Therefore, bacteriophage Qβ could not be absorbed with clay particle without coagulant. PACl could be reduce negative charge of amino acid reach to isoelectric point (non – charge amino acid). Then amino acid-RNA composition becomes non-charge and could be adsorbed with negative charge clay particle to form large aggregate.

The coagulation effectively aggregated bacteriophage Qβ to form the larger size of the aggregates and also increased coagulation time. Therefore, the aggregated bacteriophage Qβ was large enough for retained on ceramic membrane surface. This section could be conclude that coagulation at PACl dose of 3.0 mg-Al/L, 2.5 mg-Al/L and 2.0 mg-Al/L combined with ceramic membrane microfiltration were the most achievable condition for bacteriophage Qβ reduction at 1.0 μm, 0.5 μm and 0.1 pore-sized, respectively when initial bacteriophage Qβ was $8x10^6$ PFU/mL.

4.4 Reduction of bacteriophage Qβ with different initial concentration by coagulation with ceramic membrane microfiltration

In this study, to investigate the obtained reduction bacteriophage Qβ efficiency by ceramic membrane microfiltration when the initial concentration of bacteriophage Qβ in feed water was different. The different initial concentrations were $5x10^5$ PFU/mL, $4x10^6$ PFU/mL and $8x10^7$ PFU/mL. From the previous experiment, initial bacteriophage Qβ concentration was $8x10^6$ PFU/mL, the most achievable PACl dosages which completely remove bacteriophage Qβ of each ceramic membrane pore-sized (1.0 μm, 0.5 μm and 0.1) were 3.0 mg-Al/L, 2.5 mg-Al/L and 2.0 mg-Al/L, respectively. The impact of the bacteriophage Qβ concentration in spiked-surface water on the reduction efficiency by PACl coagulation combined with ceramic membrane microfiltration at the achievable PACl dosage of different pore sizes were measured as shown in Table 4.4 and figure 4.5.

Table 4.4 Reduction of different initial bacteriophage Qβ concentration

Pore sizes (μm)	The most achievable PACl dosage with microfiltration											
	0.1 (2.0 mg-Al/L**)**				**0.5 (2.5**mg-Al/L**)**				**1.0 (3.0** mg-Al/L**)**			
Initial Qβ concentration (10^6 PFU/mL)	0.5	4.0	8.0	80.0	0.5	4.0	8.0	80.0	0.5	4.0	8.0	80.0
Qβconcentration in filtrate (10^1PFU/mL)	ND	ND	ND	3.2	ND	ND	ND	4.9	ND	ND	ND	14.6
Log removal	5.7	6.6	6.9	6.4	5.7	6.6	6.9	6.2	5.7	6.6	6.9	5.7

As shown in table 4.4, the result showed that applying high bacteriophage Qβ concentration in batch experiment. Through the most achievable PACl dosage, all pore sizes exhibited up to 5 log removal efficiency at all different initial concentration. The aggregated bacteriophage Qβ were retained completely by ceramic membrane when initial concentration ranging from 10^5 to 10^6 PFU/ml. However, when the initial concentration was increased to 10^7 PFU/ml, the most achievable PACl dosage was not sufficient to reject bacteriophage Qβ by ceramic membrane filtration completely. Bacteriophage Qβ were presented in filtrated water at all pore sizes and achieved high log removal of 6.4 by 0.1 μm pore size. Similarly, the ceramic membrane filtration of 10^7 PFU/ml initial concentrations with 0.5 and 1.0 μm pore size showed low log removal of 6.2 and 5.7, respectively.

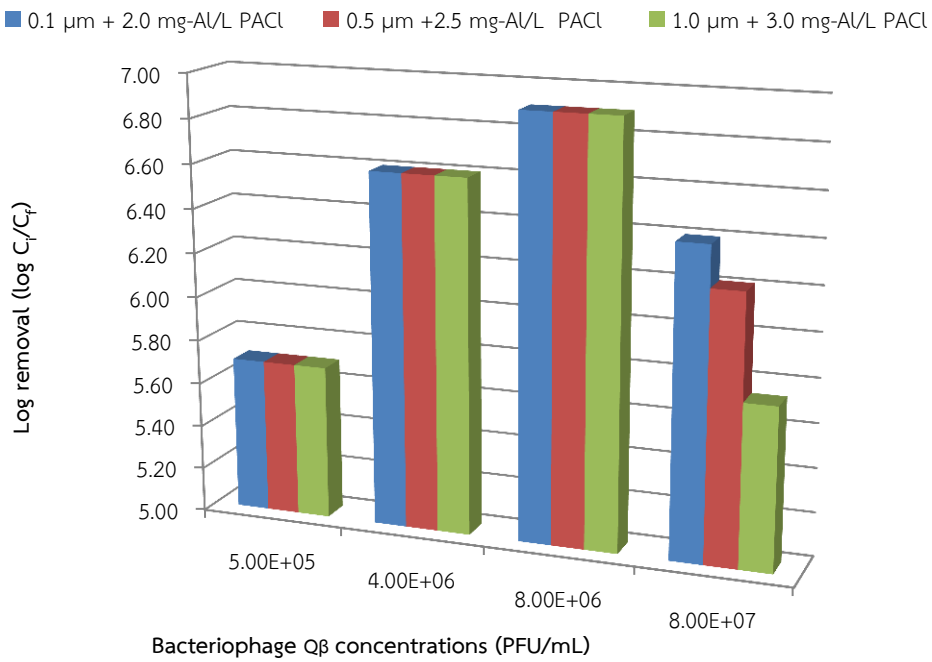


Figure 4.3 Reduction of bacteriophage Qβ with different initial concentration by coagulation with ceramic membrane microfiltration

As shown in figure 4.3, the low initial bacteriophage Qβ concentrations (5.0 $x10^5$ and $4x10^6$ PFU/mL) were totally retained bacteriophage Qβ by ceramic membrane microfiltration with coagulation and also achieved up to 5 log removal (5.7 and 6.6 log removal, respectively). High removal concentration could occur from bacteriophage Qβ aggregated with suspended solid by PACl coagulant, then the large aggregation that formed and retained on and/or in membrane surface was enhanced. In contrast, the high initial concentration required added PACl dosage to aggregate bacteriophage Qβ with clay particle and retain sufficiently by membrane filtration. Although the most achievable PACl dosage was not completely remove bacteriophage Qβ in spiked-surface water, the performance of reduction shows upper 5 log removal at all pore sizes.

The increase of initial bacteriophage Qβ concentration has an effected to the obtained reduction of bacteriophage Qβ concentration by ceramic membrane microfiltration. The highest log removal was different follow the initial bacteriophage Qβ concentration. Increasing the concentration of bacteriophage Qβ in surface water from 10^5 to 10^7 PFU/mL affected a more than 1 log drop in reduction using coagulation with ceramic membrane microfiltration. The most achievable PACl dosage from previous study could not suitable for remove all initial concentration of bacteriophage Qβ. These results related to Jacangelo *et. al*. (1995) which noticed that increasing the concentration of bacteriophage in the feed solution from 10^6 to 10^9 PFU/mL caused more than 1 log drop in removal using ultrafiltration. From this section, it's illustrated that the bacteriophage Qβ in feed water could an effect on

the obtained removal of bacteriophage Qβ by ceramic membrane microfiltration with coagulation. For the further work, which designing of bacteriophage removal experiment, the feed concentration should be constant for the different experiment if not the virus removal could be impacted.

4.5 The most achievable PACl doses of bacteriophage Qβ concentration

This section investigated the most achievable PACl dosage which used in this study to reduce bacteriophage Qβ concentration in feed water by ceramic membrane microfiltration combined with coagulation. In addition, PACl dosage was varied to find the most achievable PACl dosage for reduce bacteriophage Qβ in spiked-surface water. The bacteriophage Qβ concentration in filtrate water from the PACl coagulation combined with ceramic membrane microfiltration was measured as shown in Table 4.5.

The first initial bacteriophage Qβ concentration was $5x10^5$ PFU/ml. bacteriophage Qβ in spiked-surface water were completely retain by 0.1 μm ceramic membrane pore size at all PACl dosage and also achieved the highest log removal efficiency (5.7 log). Decreasing the concentration of bacteriophage Qβ in the feed solution from 10^6 to 10^5 PFU/mL affected 1 log drop in removal. On the other hand, bacteriophage Qβ still presented in filtrate from larger ceramic membrane pore size with small PACl doses (1.5 mg-Al/L).

To remove all bacteriophage Qβ in spiked-surface water, 0.5 μm and 1.0 μm ceramic membrane pore size required 2.0 mg-Al/L PACl dosage. The most achievable PACl dosage of 0.5 μm and 1.0 μm ceramic membrane pore size which totally removed bacteriophage Qβ and presented high log removal was 2.0 mg-Al/L PACl dosage. (Figure 4.4).

Table 4.5 Effect of bacteriophage Qβ concentration by ceramic membrane at varies PACl dosage.

Initial Qβ concentration (PFU/mL)	Pore size (μm)	coagulation PACl dosage (mg-Al/L)				
		0.0	1.5	2.0	2.5	3.0
$5x10^5$	0.1	0.56	5.7	5.7	5.7	5.7
	0.5	0.50	4.1	5.7	5.7	5.7
	1.0	0.44	3.8	5.7	5.7	5.7
$4x10^6$	0.1	0.88	6.0	6.6	6.6	6.6
	0.5	0.80	5.3	6.0	6.6	6.6
	1.0	0.75	4.2	5.0	5.9	6.6
$8x10^7$	0.1	0.20	4.6	5.5	6.3	7.9
	0.5	0.16	4.2	5.3	6.2	7.9
	1.0	0.12	3.4	4.7	5.2	6.0

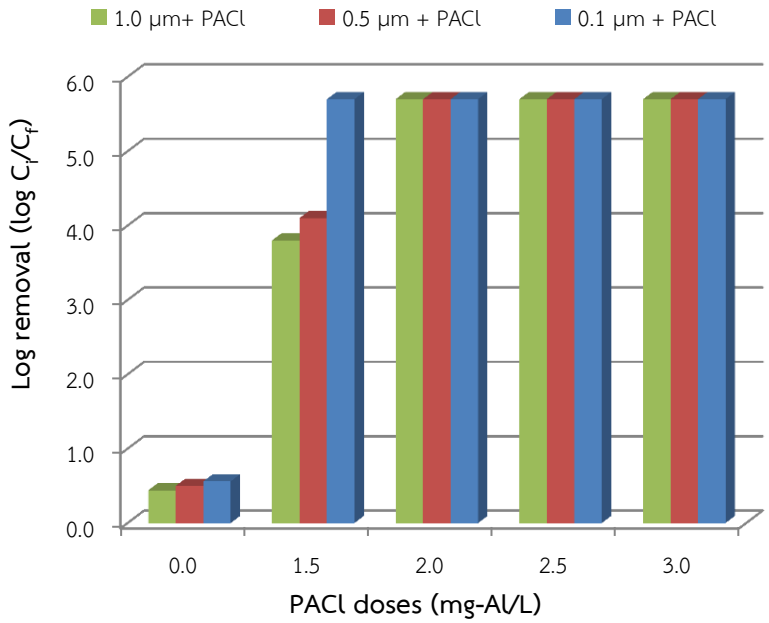


Figure 4.4 Bacteriophage Qβ log removal by coagulation combine with ceramic membrane microfiltration of 5 x10^5 PFU/mL as initial concentrations.

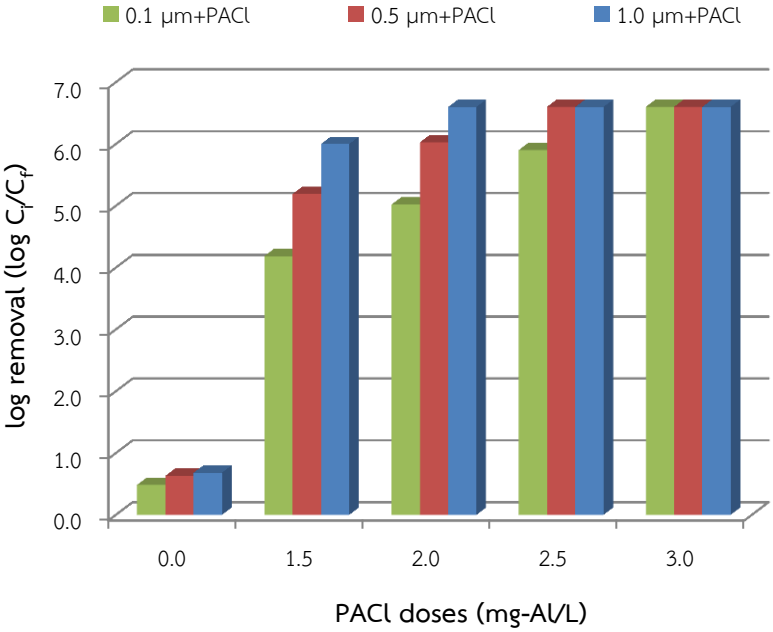


Figure 4.5 Bacteriophage Qβ log removal by coagulation combine with ceramic membrane microfiltration of 4.0x10^6 PFU/mL as initial concentrations.

Figure 4.5 shows bacteriophage Qβ log removal when initial bacteriophage Qβ concentration in spiked-surface water was 4.0 x10^6 PFU/mL. The coagulation combine with ceramic membrane microfiltration shows the 3 log higher than ceramic membrane filtration alone. At the low PACl dose (1.5 mg-Al/L), High log removal was 6.0 by 0.1 μm pore size but bacteriophage Qβ still remained in filtrated water. The combination of 0.1 μm ceramic membrane pore size with 2.0 mg-Al/L reach to the highest log removal (6.6 log), similar to the high PACl dosage(2.5 and 3 mg-Al/L). In other hand, two larger ceramic membrane pore sizes (0.5 μm and 1.0 μm) required more PACl dosage for totally removed bacteriophage Qβ with high log bacteriophage Qβ removal. The most achievable PACl dosage which removes bacteriophage Qβ completely for 0.5 μm and 1.0 μm pore sizes were 2.5 and 3.0 mg-Al/L, respectively.

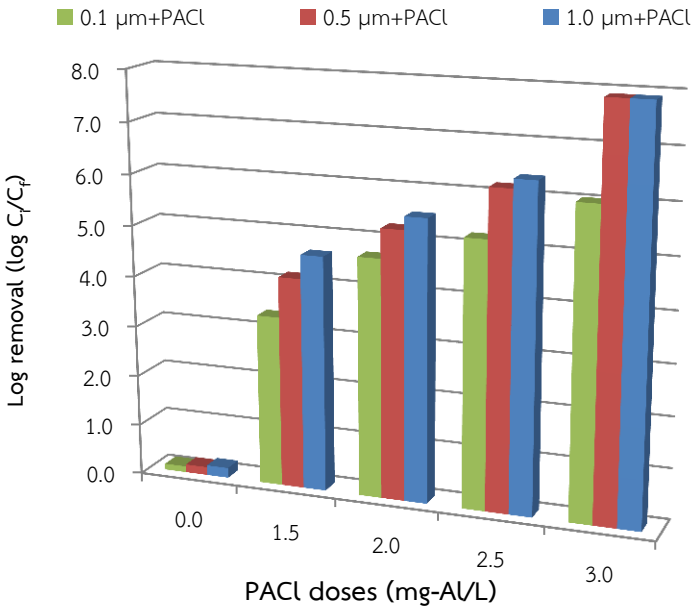


Figure 4.6 Bacteriophage Qβ log removal by coagulation combine with ceramic membrane microfiltration of 8x10^7 PFU/mL as initial concentrations.

Figure 4.6 indicated that, the removal of the high initial bacteriophage Qβ concentration was required the highest PACl dosages which use in this study. The highest log removal (7.9 log) was observed by the coagulation combined with 0.5 and 0.1 μm ceramic membrane pore sizes. At the low PACl dosing (1.5 mg-Al/L), the high log removal (4.6 log) was achieved by the coagulation with 0.1 μm ceramic membrane pore size. It indicated that the growth of aggregates was not sufficiently large to remove bacteriophage Qβ with low PACl doses at all pore size ceramic membrane with the coagulation. The high initial bacteriophage Qβ concentration required more PACl dosage for improve aggregation and removal efficiency.

Increasing the coagulant could serve to make the aggregates adequately large forms to be rejected by the ceramic membrane microfiltration. The most achievable PACl dosage for remove the high initial bacteriophage Qβ concentration was 3.0 mg-Al/L. The highest log removal was 7.9 log by 0.5 and 0.1 μm ceramic membrane pore size. Incidentally, the performances of 1.0 μm ceramic membrane pore size with the coagulation was lower than the smaller pore sizes, bacteriophage Qβ was not retained by ceramic membrane microfiltration. At 1.0 μm ceramic membrane pore size, the log removal was not improved with increasing PACl dosage. The highest PACl dosage in this study was not sufficient to form large aggregates for rejected by the ceramic membrane microfiltration and required more PACl dosage to remove bacteriophage Qβ in spiked-surface water.

4.6 Overall performance on bacteriophage Qβ by ceramic membrane microfiltration with and without coagulation

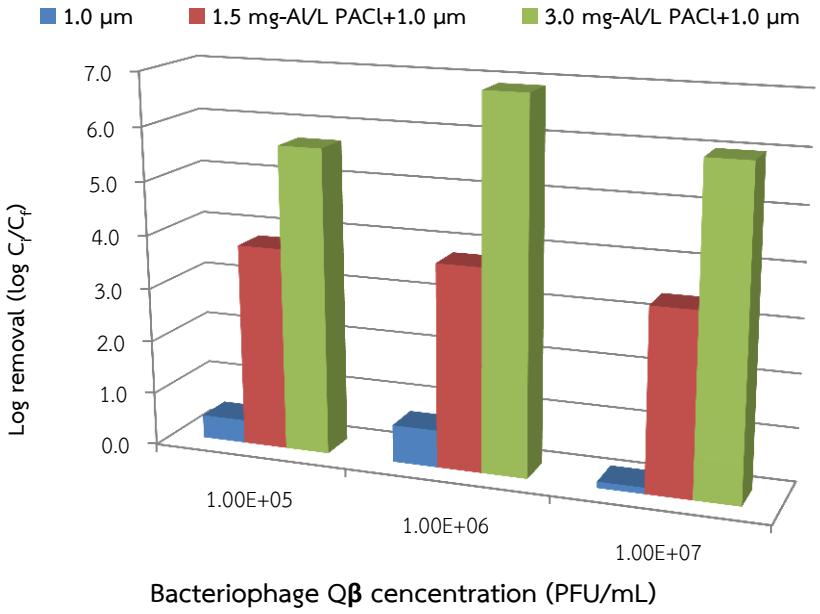
Table 4.6 shows the overall performance of bacteriophage Qβ removal by ceramic membrane microfiltration with and without coagulation. The ceramic membrane microfiltration achieved an overall removal of bacteriophage Qβ of lower than 1 log (when treating ceramic membrane filtration alone) to upper 3.0 log (when treating ceramic membrane microfiltration with coagulation). When coagulation applied, the growth of aggregated bacteriophage Qβ was larger than membrane pore size and large enough to remove by membrane filtration. However, the lowest PACl dosage (1.5 mg-Al/L) with large ceramic membrane pore size did not reduce bacteriophage Qβ as efficiency as higher dosage.

Adding PACl dosage improved bacteriophage Qβ reduction by ceramic membrane microfiltration at all pore sizes. As shown in table 4.6, the highest PACl dosage of this study achieved higher bacteriophage Qβ removal at all pore size. Especially, when applied the highest PACl dosage with the small ceramic membrane pore size. These results related to Matsui (2005) which reported that the coagulation dose thus strongly affects virus removal: the larger the coagulation dose, the greater the proportion of viruses removed.

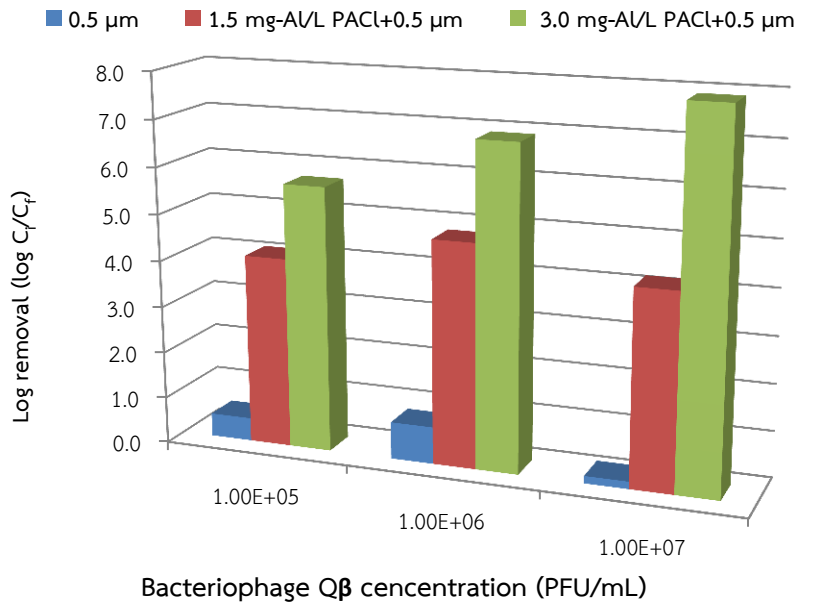
Table 4.6 Bacteriophage Qβ reduction by ceramic membrane microfiltration with and without coagulation

Filtrated water	Initial bacteriophage Qβ concentration (PFU/mL)			Log removal		
	1.00 x10^5	**1.00 x10^6**	**1.00 x10^7**	**5.7**	**6.9**	**7.9**
1.0 µm	1.80 x10^5	1.78 x10^6	6.10 x10^7	0.4	0.7	0.1
1.5 mg-Al/L PACl +1.0 µm	7.80 x10^1	5.37 x10^2	3.45 x10^4	3.8	3.8	3.4
3.0 mg-Al/L PACl +1.0 µm	1.00 x10^0	1.00 x10^0	7.60 x10^1	5.7	6.9	6.0
0.5 µm	1.57 x10^5	1.18 x10^6	5.50 x10^7	0.5	0.8	0.2
1.5 mg-Al/L PACl +0.5 µm	3.60 x10^1	3.20 x10^1	5.55 x10^3	4.1	4.8	4.2
3.0 mg-Al/L PACl +0.5 µm	1.00 x10^0	1.00 x10^0	1.00 x10^0	5.7	6.9	7.9
0.1 µm	1.38x10^5	9.33 x10^5	5.10 x10^7	0.6	0.9	0.2
1.5 mg-Al/L PACl +0.1 µm	1.00x10^0	4.57 x10^1	1.88 x10^3	5.7	5.7	4.6
3.0 mg-Al/L PACl +0.1 µm	1.00 x10^0	1.00 x10^0	1.00 x10^0	5.7	6.9	7.9

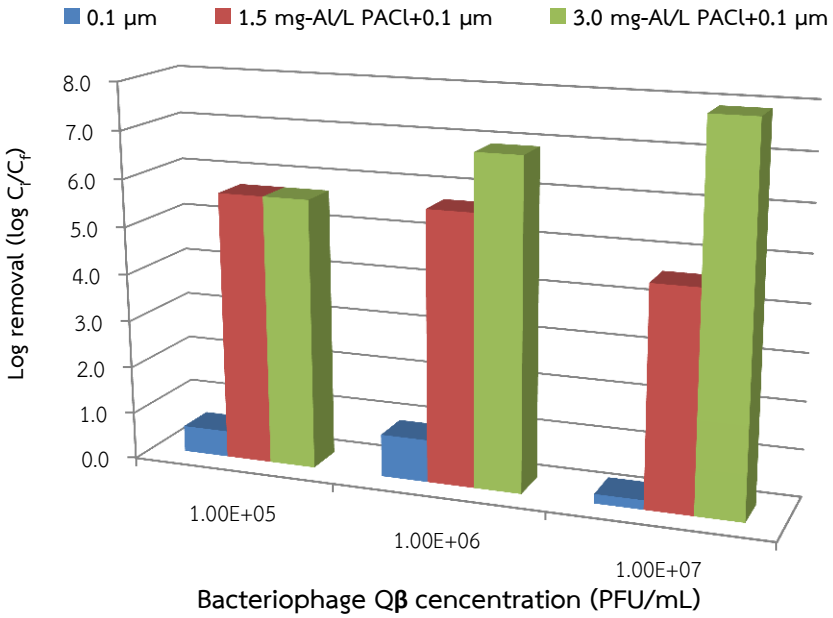
Figure 4.7 illustrated that, the operation of ceramic membrane microfiltration alone, low bacteriophage Qβ log removal was observed. However, the ceramic membrane microfiltration with coagulation was an effective barrier against bacteriophage Qβ, log removal efficiency increase.



a) Reduction of bacteriophage Qβ by 1.0 µm ceramic membrane microfiltration with and without coagulation



b) Reduction of bacteriophage Qβ by 0.5 µm ceramic membrane microfiltration with and without coagulation



c) Reduction of bacteriophage Qβ by 0.1 µm ceramic membrane microfiltration with and without coagulation

Figure 4.7 Reduction of bacteriophage Qβ by different ceramic membrane microfiltration with and without coagulation

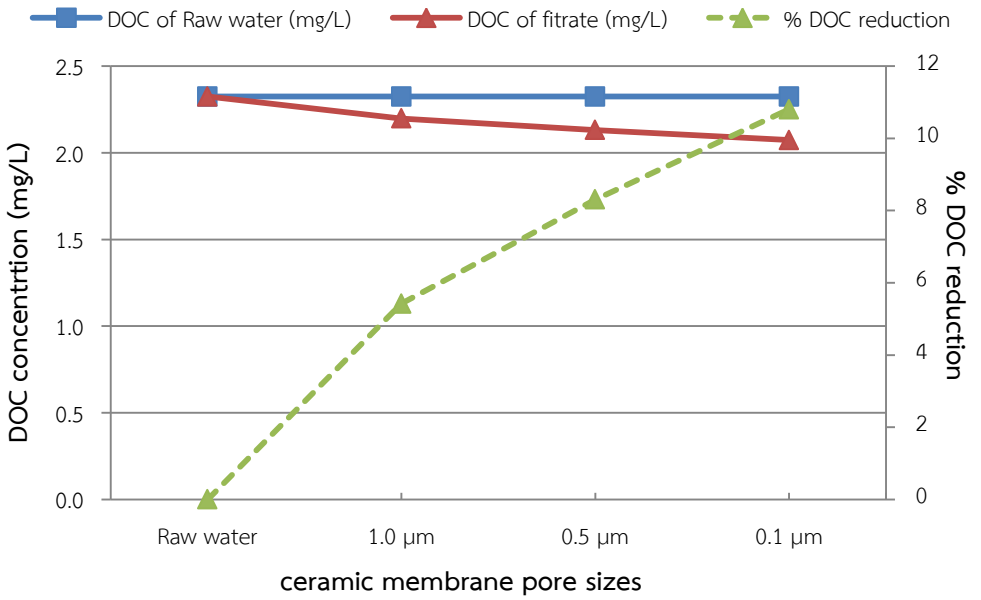
4.7 Reduction of DOM surrogate

Dissolved organic matter (DOM) is a complex mixture of hydrophilic and hydrophobic organic materials which varies in size, functional groups and reactivity (Yee et al, 2009). The several surrogate parameters must be used to describe DOM because no single surrogate parameter is capable of measuring the widely varied characteristics of DOM. Commonly surrogate parameters for DOM measurement are include dissolved organic carbon (DOC), ultraviolet absorbance at wavelength of 254 nm (UV-254), specific Ultraviolet Absorbance (SUVA), which were observed in this study.

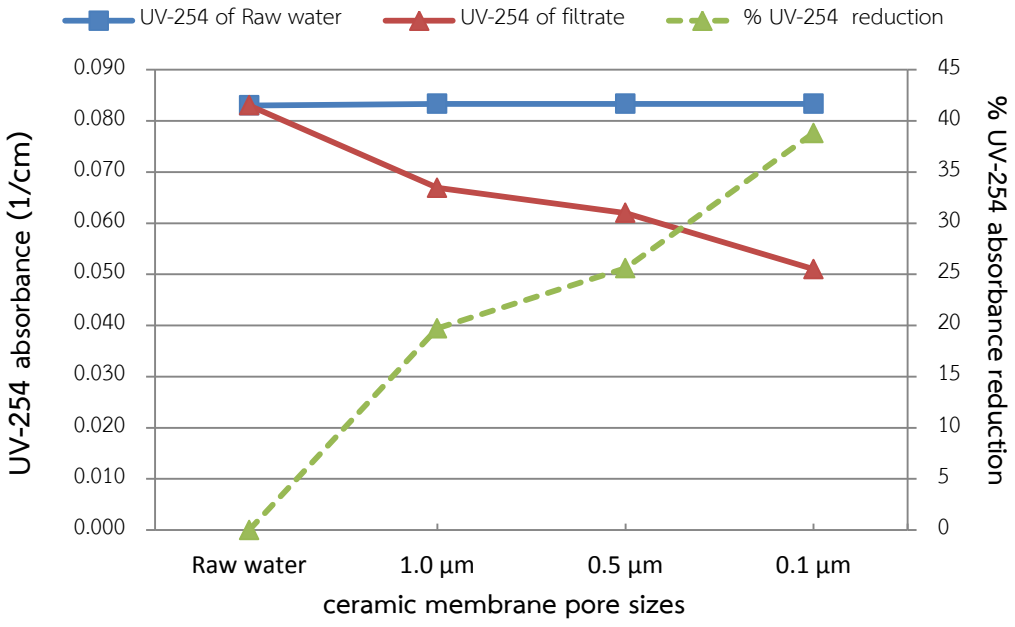
4.7.1 Reduction of DOC, UV-254 and SUVA by ceramic membrane microfiltration

As can be seen in Figure 4.8 (a), The DOC concentration were reduced from 2.324 mg/L in raw surface water to 2.198, 2.131 and 2.073 mg/L by 1.0μm, 0.5μm and 0.1μm ceramic membrane pore sizes, respectively. Percent DOC reductions were 5.42%, 8.30% and 10.80% at 1.0μm, 0.5μm and 0.1μm ceramic membrane pore sizes, respectively. From the result, it can be indicated that the percent DOC reduction were increased by the smaller pore size. According to these very low DOC removal results obtained, it can be stated that the efficiency of the 1.0μm, 0.5μm and 0.1μm ceramic membrane microfiltration relies on the sieving mechanism alone could not be sufficient to reduce DOC concentration.

The results of UV-254 absorbance reduction in figure 4.10(b) showed that the ceramic membrane microfiltration can reduce UV-254 absorbance in raw surface water from 0.083 cm^{-1} to 0.067, 0.062 and 0.051 cm^{-1} by 1.0μm, 0.5μm and 0.1μm ceramic membrane pore sizes, respectively. Percent UV-254 reductions were 19.72%, 25.6% and 38.8% by 1.0μm, 0.5μm and 0.1μm ceramic membrane pore sizes, respectively. The UV-254 absorbance was used to indicate the aromatic hydrocarbon in water. From the results, low UV-254 reductions were obtained but greater than DOC reduction at the same ceramic membrane pore sizes. In fact, the value of DOC in water used to indicate the aromatic and aliphatic hydrocarbons in water. It can be stated that ceramic membrane microfiltration has capable to reduce aromatic hydrocarbon in water.



a) Residual DOC concentration and percent DOC reduction by ceramic membrane microfiltration



b) Residual UV-254 and percent UV-254 reduction by ceramic membrane microfiltration

Figure 4.8 Reduction and percent reduction of DOC and UV-254 absorbance by ceramic membrane microfiltration

SUVA was used as an index of humic content in water (Edzwald, 1993), calculated from the ratio between UV absorbance wavelength 254 nm to dissolved organic carbon (DOC) concentration. In addition, the SUVA values can be used as an indicator of coagulation ability to remove organic matter. The result of SUVA values of spiked surface water by ceramic membrane microfiltration are shown in Figure 4.9.

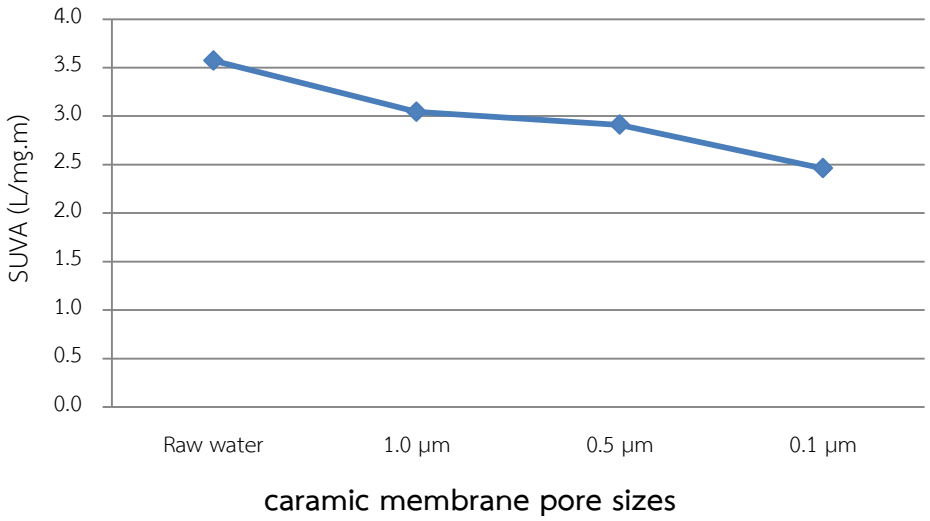


Figure 4.9 SUVA values of ceramic membrane microfiltration at various pore sizes

The result of SUVA values of ceramic membrane microfiltration show the reduction of SUVA from 3.571 (L/mg-m) to 3.044, 2.909 and 2.460 L/mg-m by lager to small ceramic membrane pore sizes. The small pore size shows higher decreased SUVA values than lager and presented SUVA values under 3 L/mg-m. As Edzwald and Van Benschoten (1985) reported that, SUVA values of less than 3 L/mg-m signify water containing mostly non-humic material, low in average molecular weight and difficult to remove by coagulation. On the other hand, SUVA values of 4-5 L/mg-m are typical of waters containing primarily humic material. SUVA of humic sample

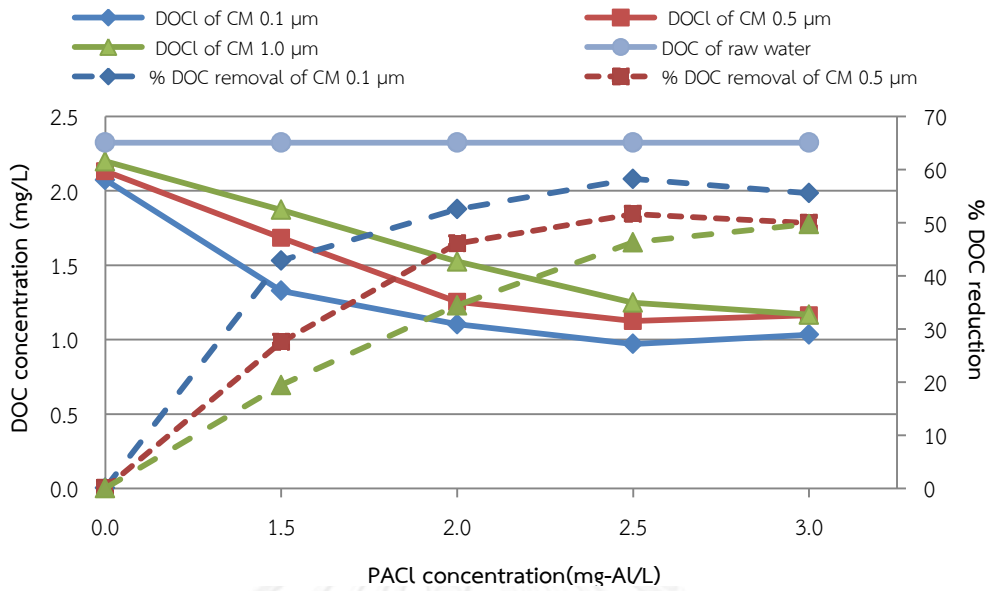
depends on the molecular weight of substances. The high of SUVA tend to indicate high humic content (Petterson et al., 1995) and more readily removed by coagulation. Form the results, it can be stated that SUVA were removed to under 3 L/mg-m by small pore size ceramic membrane microfiltration, can reduce mostly humic material in spiked-surface water.

4.7.2 Reduction of DOC, UV-254 and SUVA by ceramic membrane filtration with coagulation

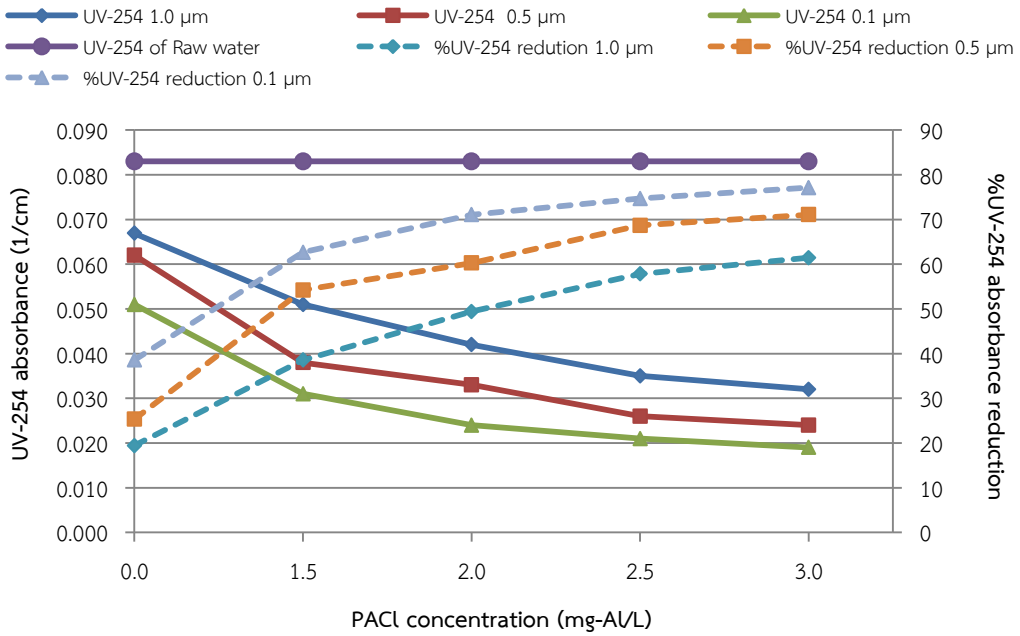
The reduction of DOC and UV-254 reduction by ceramic membrane microfiltration combined with coagulation are shown in Figure 4.10(a). The results shows that the DOC concentration was reduced from 2.324 mg/L in raw surface water to 1.872, 1.524, 1,248 and 1.168 by 1.0 μm pore size ceramic membrane microfiltration, 1.684, 1.253, 1.124 and 1.163 by 0.5 μm pore size ceramic membrane microfiltration, 1.328, 1.103, 0.971 and 1.033 by 0.1 μm pore size ceramic membrane microfiltration at PACl dosage 1.5, 2.0, 2.5 and 3.0 mg-Al/L, respectively. Percent DOC reductions when coagulation combined were higher than only ceramic membrane microfiltration. The highest percent DOC reduction of spiked-surface water was 58.22% by coagulation combined with 0.1 μm at PACl dose 2.5 mg-Al/L. The percent DOC reductions were low with large pore sizes. It could be stated that the amount of PACl was not enough to eliminate DOC and the size of coagulated compound quiet smaller than pore size of ceramic membrane. Therefore DOC quiet current in spiked-surface water.

From the previous results, the large pore size also demonstrates low reduction of bacteriophage Qβ concentration. It can be stated that low PACl dosage with large pore size ceramic membrane was not sufficient to reduce bacteriophage Qβ and DOC concentration. DOC reduction of spiked-surface water by coagulation combined with 1.0 μm with various PACl dosages could reduce DOC concentration in range 19.45%- 34.42% and 46.30% - 49.74% reduction. The results obtained were lower than 50% DOC removal. This could be implied that coagulation combined with 1.0 μm was inadequate condition to remove DOC from spiked-surface water.

The results of UV-254 absorbance reduction in figure 4.10(b) showed that the ceramic membrane micofiltration with coagulation could reduce UV-254 absorbance in raw surface water from 0.083 cm-1 to 0.051, 0.042, 0.035 and 0.032 cm-1 by 1.0 μm pore size ceramic membrane microfiltration, 0.038, 0.033, 0.026 and 0.024 cm-1 by 0.5 μm pore size ceramic membrane microfiltration, 0.031, 0.024, 0.021 and 0.019 cm-1 by 0.1 μm pore size ceramic membrane microfiltration at PACl dosage 1.5, 2.0, 2.5 and 3.0 mg-Al/L, respectively. Percent UV-254 reductions were 38.5-61.4%, 54.2-71.1% and 62.5-77.01% by 1.0μm, 0.5μm and 0.1μm ceramic membrane pore sizes, respectively. The UV-254 reduction performance shows upper 50% reduction at all PACl dasage and pore size of ceramic membrane. Adding i coagulation could increase percent reduction of UV-254. As stated previously, The UV-254 absorbance was used to indicate the aromatic hydrocarbon in water. From the results, it could be stated that the coagulation combined with ceramic membrane was increase UV-254 reductions by reduce aromatic hydrocarbon in water, higher than ceramic membrane microfiltration alone.



a) DOC concentration and percent DOC reduction by ceramic membrane microfiltration with coagulation



b) UV and percent UV reduction by ceramic membrane microfiltration with coagulation

Figure 4.10 Reduction and percent reduction of DOC and UV-254 absorbance by ceramic membrane microfiltration with coagulation

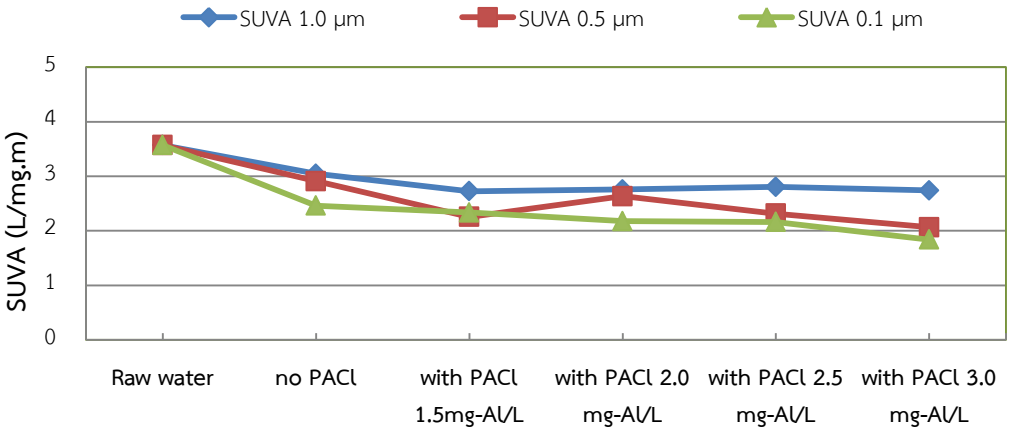


Figure 4.11 SUVA values of filtrate by ceramic membrane microfiltration with coagulation

As shown in Figure 4.11, the PACl coagulation with ceramic membrane microfiltration could reduce SUVA from spiked-surface water by show under 3 L/mg-m at all pore sizes. The result of SUVA values of ceramic membrane microfiltration show the reduction of SUVA from 3.571 (L/mg-m) to 2.724, 2.257 and 2.334 L/mg-m by the lowest PACl dosage coagulation with ceramic membrane microfiltration. The small pore size shows higher decreased SUVA values than lager and presented SUVA values under 3 L/mg-m. As stated previously, water that having low SUVA (<3 L/mg-m) has been found to have organic matter mostly in term of non-humic in character. The combination of ceramic membrane microfiltration with the coagulation could decrease organic matter mostly in term of humic-like in character.

Similarly to this section, the low DOC reduction were obtained in all experiment when used ceramic membrane microfiltration alone, it can described that the only sieving mechanism of 1.0 µm, 0.5 µm and 0.1 µm ceramic membrane

microfiltration could not be sufficient to reduce DOC in spiked-river water. When compared with the PACl coagulation membrane microfiltration. It was assumed that DOC removal by PACl coagulation at all PACl doses with ceramic membrane microfiltration were significant higher than that by ceramic membrane microfiltration alone. The PACl coagulation may increase the performance of ceramic membrane microfiltration by increase the detention time of flocs formation inside 8 meters-nylon tube prior to form the larger flocs size than pore size of ceramic membrane. The percent reduction was upper than 40 percent reduction when applied 2.5 and 3.0 mg-Al/L PACl dosages at all ceramic membrane microfiltration. The highest reduction (58.2%) was present when applied 3.0 mg-Al/L PACl dosages with 0.1 μm ceramic membrane microfiltration. Enhanced coagulation according to USEPA (1998), the DOC in raw water a between less than 2.0-4.0 mg/L and alkalinity of about 0-60 mg/L as $CaCO_3$, the water treatment process was required to remove 40 percent of DOC.

The average SUVA values observed was 3.58 L/mg-m of Ping River water. It can be stated that Ping River water mostly contains humic material. The moderate UV-254 removal was by coagulation with 3.0 mg- Al/l combined with 0.1 μm that exhibited the percent removal about 70%.

4.8 Reduction of bacteria by ceramic membrane microfiltration with and without coagulation

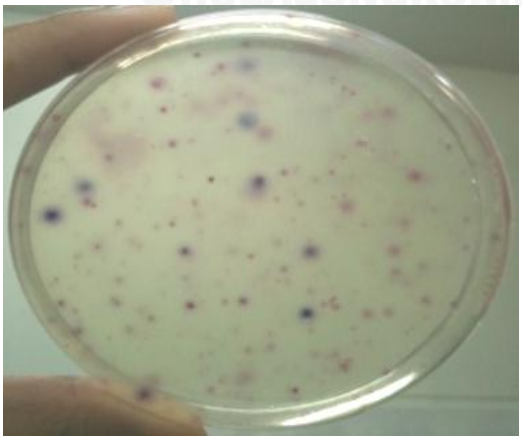
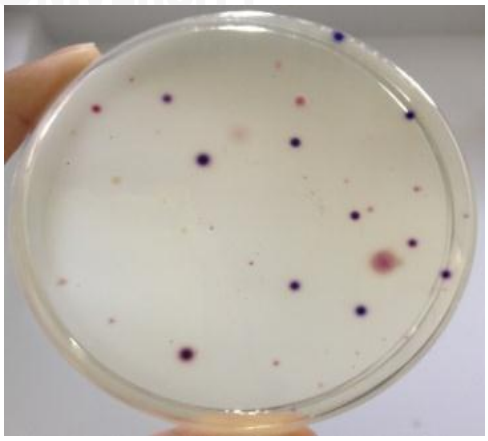
Total and fecal coliforms have been used extensively for many years as indicators for determining the sanitary quality of natural water. This section was evaluating water quality after pass through the ceramic membrane microfiltration with and without coagulation using total coliform and E. coli as microbial indicators.

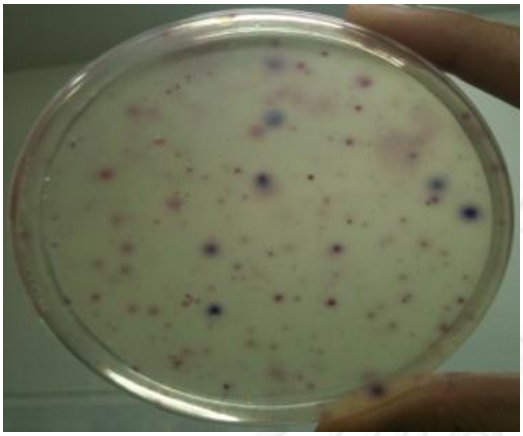
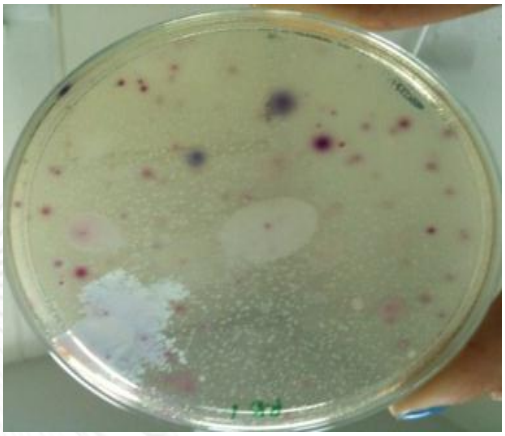
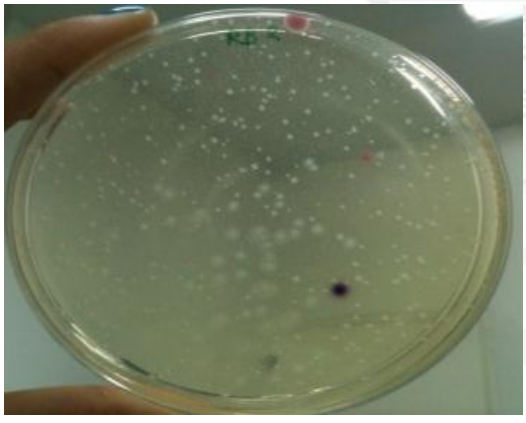
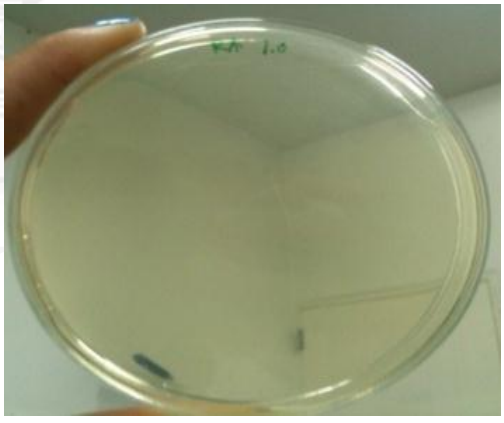
In order to investigate the fecal pollution removal efficiency of ceramic membrane microfiltration, total coliform and *E. coli* were detected from the filtrated of spiked-surface waters. Total coliform and E. coli detection were analyzed by single agar layer method using Chromocult Coliform agar as culture media. Triple analyzed plate counts were always done in each dilution. Salmon to red colonies and dark-blue to violet colonies were counted as total coliforms. Dark-blue to violet colonies were counted as *E.coli*. The concentration of microbes was reported as Colonies Forming Unit/ ml (CFU/ml). The detection limit in this experiment was 1 CFU/ml. The example of Total coliform and *E.coli* from filtrated water was shown in Table 4.7.

In this experiment, Total coliform and *E.coli* was found from Ping River water in amount of 296±1.52 CFU/mL and 27±0.05 CFU/ml, respectively. The results obtained that Only 1.0 μm ceramic membrane microfiltration and when applied with coagulation at lowest PACl dosage could fairly remove total coliform. However, *E.coli* was completely removed by 1.0 μm ceramic membrane microfiltration with coagulation at lowest PACl dosage. On the other hand, the small pore sizes (0.5 μm

and 0.1 μm) ceramic membrane microfiltration could remove coliform and *E.coli* completely similarly to treating with ceramic membrane microfiltration with coagulation combined at lowest PACl dosage. According to the results, it could be concluded that without treating process the microbial quality of the water sources was poor and unacceptable for human consumption due to faecal pollution (DWAF, 1998 set the maximum limit for no risk of faecal coliforms is 0 CFU/100ml). It could be certainly suggested that the ceramic membrane microfiltration with and without coagulation could be efficiently used for feacal pollution treatment; since, coliform bacteria are larger than the absolute pore size of the ceramic membranes (0.6–1.2 μm in diameter by 2–3 μm in length).

Table 4.7 Total coliform and *E.coli* from filtrated Ping River water by 0.1 μm ceramic membrane microfiltration and 0.1 μm ceramic membrane microfiltration with 2.5mg-Al /L PAC dosage.

Ping river water	
Total Coliform = 29.6 x10^{1}CFU/mL.	*E. coli* = 27 x10^{0} CFU/mL.
	

<table>
<tr><td colspan="2">1.0 µm</td></tr>
<tr><td>Total Coliform = 15x10^{0}CFU/mL.</td><td><i>E. coli</i> =4.6x10^{0} CFU/mL.</td></tr>
<tr><td>  </td><td>  </td></tr>
<tr><td colspan="2">2.5mg-Al /L + 1.0 µm</td></tr>
<tr><td>Total Coliform =2x10^{0} CFU/mL.</td><td>E. coli = -</td></tr>
<tr><td>  </td><td>  </td></tr>
</table>

CHAPTER V

CONCLUSIONS

Based on the obtained results from the study of bacteriophage Qβ reduction by different ceramic membrane pore size, reduction of DOM surrogate parameters (DOC, UV-254, and SUVA), filtrated water by PACl coagulation combined with ceramic membrane microfiltration of Ping River water, the following conclusions could be drawn.

The pore size of ceramic membrane was not affected bacteriophage Qβ removal. Ceramic membrane microfiltration with pore size larger than 0.1 μm showed lower log removal, could not act as physical barrier to bacteriophage Qβ. The ceramic membrane pore sizes of 0.1, 0.5 and 1.0 μm could remove bacteriophage Qβ about 0.9 log, 0.8 log and 0.7 log, respectively. Only ceramic membrane microfiltration cannot remove virus in wastewater alone.

The PACl coagulations have a strongly effect to bacteriophage Qβ removal. The most achievable PACl dosage for 0.1, 0.5 and 1.0 μm pore sizes were 2.0, 2.5 and 3.0 mg-Al/L, respectively. The ceramic membrane microfiltration with PACl coagulation at the most achievable PACl dosage achieved 6.9 log removals. The application of coagulation can develop virus removal efficiency of ceramic

membrane microfiltration. Coagulation processes help to aggregate small particles in water to larger aggregates.

The bacteriophage Qβ removals by ceramic membrane microfiltration with PACl coagulation with the low initial bacteriophage Qβ concentrations (5.0 $x10^5$ PFU/mL) were completely retained by all pore size. The bacteriophage Qβ still presented in filtrated water when the initial bacteriophage Qβ concentrations were high (1.0 $x10^7$ PFU/mL). The highest PACl (3.0 mg-Al/L) was not sufficiently aggregate and remove by 1.0 μm ceramic membrane microfiltration with the PACl coagulation. The high initial bacteriophage Qβ required more PACl dosage for improve aggregation and removal efficiency by 1.0 μm ceramic membrane microfiltration.

The high initial bacteriophage Qβ concentration (1.0 $x10^7$ PFU/mL) were completely retained and achieved 7.9 log g at the highest PACl dosage which use in this study (3.0 mg-Al/L) with 0.1μm and 0.5μm ceramic membrane microfiltration. Thus, the PACl coagulation with 0.5 μm ceramic membrane microfiltration was the suitable condition for reduce bacteriophage Qβ since it can produce in larger filtrated volume and also reduces the cost of producing drinking water when actual operations as well. The high removal efficiency could occurs by increase amount of PACl, extending the coagulation time, the smallest pore size are not necessary.

The low DOC reduction were obtained in all experiment when used ceramic membrane microfiltration alone, that the only sieving mechanism ceramic membrane

microfiltrations could not be sufficient to reduce DOC in water. The PACl coagulation may increase the performance of ceramic membrane filtration by increase the detention time of flocs formation inside 8 meters-nylon tube prior to form the larger flocs size than pore size of ceramic membrane. The highest percent DOC reduction of spiked-surface water was 58.22% by coagulation combined with 0.1 μm at PACl dose 2.5 mg-Al/L.

Filtrated water through the ceramic membrane microfiltration with coagulation can be used in human activities safely due to the virus was remove. Not only virus was removed by ceramic membrane microfiltration with coagulation, the others microbial were removing as well. Total coliform and *E.coli* were used as indicators for determining the faecal pollution reduction in this experiment. Total coliform and *E.coli* was found from Ping River water in amount of 296±1.52 CFU/mL and 27±0.05 CFU/mL, respectively. The results obtained from 1.0 μm ceramic membrane microfiltration with the lowest PACl dosage (1.5 mg-Al /L) coagulation and 1.0 μm ceramic membrane microfiltration alone could fairly remove total coliform. In contrast, 0.5 μm and 0.1μm ceramic membrane microfiltration and ceramic membrane microfiltration combined with coagulation could remove total coliform and *E.coli* completely. The microbial quality of the water sources was acceptable for human consumption due to fecal pollution completely remove (DWAF, 1998 set the maximum limit for no risk of faecal coliforms is 0 CFU/100mL).

CHAPTER VI

RECOMMENDATIONS FOR FUTURE WORK

The following statements are recommended for future studies.

1. Most surface water treatment plants use aluminum in the form of alum (aluminum sulphate) to help remove harmful waterborne microorganisms and other particles by causing them to clump together (coagulate) into larger particles that are then easily removed by sedimentation and filtration. Aluminum can become poisonous and have a range of health effects from skeletal deformities to brain degeneration. Thus, the intake of aluminum in drinking water generally amounts to less than 5% of the total daily intake for an adult. In this case, the amount of aluminum in the filtrate should be investigated to confirm that the amount of aluminum residue in the filtrate does not exceed the standards and will not affect to human health.

2. The contact time of coagulation should be developed by increasing the length of tube for increasing the detention time of PACl coagulation in order to make the comparison with the results obtained in this study to achieve the suitable condition for bacteriophage Qβ reduction.

3. Run time of PACl coagulation with ceramic membrane is interesting for evaluate the effect of aggregation on the membrane surface. If Coagulation time affected virus removal in the coagulation-microfiltration hybrid system (Matsushita *et al*., 2004) the longer coagulation time can developed the reduction in virus removal.

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APPENDIX

APPENDIX A

DOC, UV-254, SUVA, TURBIDITY, ALKALINITY, TEMPERATURE, AND pH

Table A-1: DOC, UV-254, SUVA, Turbidity, Alkalinity, Temperature and pH of raw River water

pH	Temperature (°C)	Alkalinity (mg/L as $CaCO_3$)	Turbidity (NTU)	DOC (mg/L)	UV-254 (cm^{-1})	SUVA (L/mg-m)
7.69±02	24.7±0.25	222±4.72	41.77±44	2.324±0.03	0.086.001	3.58±0.09

Table A-2: DOC, UV-254, SUVA, Turbidity, Alkalinity, Temperature and pH of filtrated water by ceramic membrane microfiltration.

Ceramic membrane Pore sizes (µm)	pH	Temperature (°C)	Alkalinity (mg/L as $CaCO_3$)	Turbidity (NTU)	DOC (mg/L)	UV-254 (cm^{-1})	SUVA (L/mg-m)
1.0	7.77	22.8	119.6	0.71	2.198	0.067	3.044
0.5	7.78	22.7	115.0	0.07	2.131	0.062	2.909
0.1	7.77	22.5	110.4	0.04	2.073	0.051	2.460

Table A-3: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water at initial bacteriophage Qβ concentration 8.00×10^6 PFU/mL and filtrated water by 1.0 μm ceramic membrane microfiltration with various PACl concentration.

Parameters	PACl dosage (mg-Al/L)				
	0.0	**1.5**	**2.0**	**2.5**	**3.0**
Turbidity (NTU)	41.77	0.17	0.05	<0.01	<0.01
Temperature (°C)	24.7	24.4	22.4	24.7	25.4
pH	7.69	7.43	7.36	7.34	7.32
EC (μs/cm)	222	224	226	233	251
DOC (mg/L)	1.790	1.328	1.103	0.971	1.033
Alkalinity (mg/L as $CaCO_3$)	108.8	124.2	119.6	124.2	128.8
UV-254 (cm^{-1})	0.083	0.031	0.024	0.021	0.019
SUVA (L/mg.m)	3.92	2.33	2.17	2.16	1.83

Table A-4: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water at initial bacteriophage Qβ concentration 8.00×10^6 PFU/mL and filtrated water by 0.5 μm ceramic membrane microfiltration with various PACl concentration.

Parameters	PACl dosage (mg-Al/L)				
	0.0	**1.5**	**2.0**	**2.5**	**3.0**
Turbidity (NTU)	41.77	0.11	0.02	<0.01	<0.01
Temperature (°C)	24.7	22.1	22.5	22.4	21.7
pH	7.69	7.53	7.51	7.3	7.76
EC (μs/cm)	222	239	238	248	215
DOC (mg/L)	1.790	1.684	1.253	1.124	1.163
Alkalinity (mg/L as $CaCO_3$)	108.8	115.0	124.2	119.6	119.6
UV-254(cm^{-1})	0.083	0.038	0.033	0.026	0.024
SUVA (L/mg.m)	3.92	2.25	2.63	2.31	2.06

Table A-5: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water at initial bacteriophage Qβ concentration $8.00x10^6$ PFU/mL and filtrated water by 0.1 μm ceramic membrane microfiltration with various PACl concentration.

Parameters	PACl dosage (mg-Al/L)				
	0.0	1.5	2.0	2.5	3.0
Turbidity (NTU)	41.77	3.03	3.48	3.39	3.88
Temperature (°C)	24.7	23.2	23.3	23.1	23.4
pH	7.69	7.85	7.93	7.97	7.92
EC (μs/cm)	222	242	245	245	233
DOC (mg/L)	1.790	1.872	1.524	1.248	1.168
Alkalinity (mg/L as $CaCO_3$)	108.8	117.6	109.2	128.8	138
UV-254(cm^{-1})	0.083	0.051	0.042	0.035	0.032
SUVA (L/mg.m)	3.92	2.724	2.756	2.804	2.740

Table A-6: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of filtrated water by 0.1 μm ceramic membrane microfiltration with 2.0 mg-Al/L PACl coagulation at various initial concentration of bacteriophage Qβ.

Parameters	0.1 μm ceramic membrane + 2.0 mg-Al/LPACl			
bacteriophages concentration (PFU/mL)	$5.00x10^5$	$4.00x10^6$	$8.00x10^6$	$8.00x10^7$
Turbidity (NTU)	<0.01	<0.01	<0.01	<0.01
Temperature (°C)	21.4	22.3	17.5	18.2
pH	7.76	7.57	7.58	7.49
EC (μs/cm)	224	238	248	261
DOC (mg/L)	1.163	1.253	1.107	1.351
Alkalinity (mg/L as $CaCO_3$)	115.0	124.2	124.2	133.4
UV-254(cm^{-1})	0.027	0.059	0.064	0.083
SUVA (L/mg.m)	2.32	4.70	5.78	6.14

Table A-7: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of filtrated water by 0.5 μm ceramic membrane microfiltration with 2.5 mg-Al/L PACl coagulation at various initial concentration of bacteriophage Qβ.

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Parameters	0.5μm ceramic membrane+2.5 mg-Al/L PACl			
bacteriophages concentration (PFU/mL)	$5.00x10^5$	$4.00x10^6$	$8.00x10^6$	$8.00x10^7$
Turbidity (NTU)	<0.01	<0.01	<0.01	<0.01
Temperature (°C)	22.2	21.7	21.6	21.6
pH	7.23	7.25	7.28	7.47
EC (μs/cm)	226	225	251	272
DOC (mg/L)	1.175	1.176	1.126	1.249
Alkalinity (mg/L as CaCO3)	128.8	128.8	128.8	133.4
UV-254(cm^{-1})	0.026	0.068	0.073	0.076
SUVA (L/mg.m)	2.21	5.78	6.48	6.08

Table A-8: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of filtrated water by 1.0 μm ceramic membrane microfiltration with 3.0 mg-Al/L PACl coagulation at various initial concentration of bacteriophage Qβ.

Parameters	1.0 μm ceramic membrane+ 3.0 mg-Al/LPACl			
bacteriophages concentration (PFU/mL)	$5.00x10^5$	$4.00x10^6$	$8.00x10^6$	$8.00x10^7$
Turbidity (NTU)	0.31	0.31	0.38	0.18
Temperature (°C)	17.2	17.6	18.6	22.2
pH	7.4	7.36	7.4	7.46
EC (μs/cm)	230	242	248	289
DOC (mg/L)	1.167	1.145	1.172	1.205
Alkalinity mg/L as CaCO3)	115.0	115.0	115.0	115.0
UV-254(cm^{-1})	0.033	0.048	0.058	0.065
SUVA (L/mg.m)	2.82	4.19	4.94	5.39

Table A-9: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water with initial bacteriophage Qβ concentration at 5 x 10^5 PFU/mL and filtrated water by 1.0 μm ceramic membrane microfiltration at various PACl concentration.

Parameters	PACl dosage (mg-Al/L)				
	0.0	1.5	2.0	2.5	3.0
Turbidity (NTU)	0.93	1.17	0.91	1.08	0.86
Temperature (°C)	22.5	26.6	26.2	26.8	26.7
pH	7.68	7.69	7.56	7.53	7.45
EC (μs/cm)	243	264	261	261	262
DOC (mg/L)	2.167	1.708	1.363	1.261	1.167
Alkalinity (mg/L as CaCO3)	110.4	124.2	124.2	128.8	124.2
UV-254(cm^{-1})	0.054	0.039	0.027	0.035	0.028
SUVA (L/mg.m)	2.49	2.28	1.98	2.77	2.39

Table A-10: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water with initial bacteriophage Qβ concentration at 5 x 10^5 PFU/mL and filtrated water by 0.5 μm ceramic membrane microfiltration at various PACl concentration.

Parameters	PACl dosage (mg-Al/L)				
	0.0	1.5	2.0	2.5	3.0
Turbidity (NTU)	0.14	0.05	0.12	0.05	0.05
Temperature (°C)	23.8	21.8	22.4	20.7	19.4
pH	7.7	7.38	7.19	7.19	7.28
EC (μs/cm)	239	255	270	276	287
DOC (mg/L)	2.145	1.508	1.283	1.175	1.108
Alkalinity (mg/L as CaCO3)	115.0	147.5	144.5	147.5	142.5
UV-254(cm^{-1})	0.051	0.033	0.036	0.027	0.029
SUVA (L/mg.m)	2.3	2.18	2.80	2.29	2.61

Table A-11: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water with initial bacteriophage Qβ concentration at 5 x 10^5 PFU/mL and filtrated water by 0.1 μm ceramic membrane microfiltration at various PACl concentration.

Parameters	PACl dosage (mg-Al/L)				
	0.0	**1.5**	**2.0**	**2.5**	**3.0**
Turbidity (NTU)	0.09	<0.01	<0.01	<0.01	0.08
Temperature (°C)	24.3	28.5	27.9	27.6	27.6
pH	7.69	7.89	7.63	7.59	7.57
EC (μs/cm)	243	268	278.5	270	258
DOC (mg/L)	2.108	1.327	1.158	1.105	1.119
Alkalinity (mg/L as CaCO3)	108.8	136.0	124.2	128.8	124.2
UV-254(cm^{-1})	0.053	0.034	0.033	0.037	0.024
SUVA (L/mg.m)	2.51	2.56	2.84	3.34	2.14

Table A-12: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water with initial bacteriophage Qβ concentration at 4 x 10^6 PFU/mL and filtrated water by 1.0 μmceramic membrane microfiltration at various PACl concentration.

Parameters	PACl dosage (mg-Al/L)				
	0.0	**1.5**	**2.0**	**2.5**	**3.0**
Turbidity (NTU)	2.07	1.09	0.61	0.16	0.09
Temperature (°C)	23.4	26.6	27	26.4	22.1
pH	7.68	7.44	7.21	7.32	7.16
EC (μs/cm)	238	220	220	216	214
DOC (mg/L)	2.184	1.582	1.332	1.132	1.145
Alkalinity (mg/L as CaCO3)	119.0	115.0	115.0	115.0	126.5
UV-254(cm^{-1})	0.052	0.026	0.038	0.026	0.019
SUVA (L/mg.m)	2.39	1.64	2.85	2.29	1.65

Table A-13: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water with initial bacteriophage Qβ concentration at 4 x 10^6 PFU/mL and filtrated water by 0.5 μm ceramic membrane microfiltration at various PACl concentration.

Parameters	PACl dosage (mg-Al/L)				
	0.0	**1.5**	**2.0**	**2.5**	**3.0**
Turbidity (NTU)	0.97	0.07	0.14	<0.01	0.03
Temperature (°C)	22.7	27.5	25.5	24.7	21.9
pH	7.7	7.6	7.5	7.6	7.5
EC (μs/cm)	249	217	219	223	227
DOC (mg/L)	2.073	1.545	1.364	1.176	1.109
Alkalinity (mg/L as CaCO3)	110.0	115.0	115.0	110.4	115
UV-254(cm^{-1})	0.054	0.029	0.031	0.037	0.029
SUVA (L/mg.m)	2.61	1.87	2.27	3.14	2.61

Table A-14: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water with initial bacteriophage Qβ concentration at 4 x 10^6 PFU/mL and filtrated water by 0.1 μm ceramic membrane microfiltration at various PACl concentratioin.

Parameters	PACl dosage (mg-Al/L)				
	0.0	**1.5**	**2.0**	**2.5**	**3.0**
Turbidity (NTU)	0.61	0.11	<0.01	<0.01	<0.01
Temperature (°C)	24.9	17.3	19.2	20.1	20
pH	7.71	7.72	7.45	7.53	7.51
EC (μs/cm)	251	238	220	226	219
DOC (mg/L)	2.035	1.442	1.253	1.016	1.142
Alkalinity (mg/L as CaCO3)	115.0	132.5	152.5	126.5	149.5
UV-254(cm^{-1})	0.053	0.039	0.0330	0.035	0.024
SUVA (L/mg.m)	2.63	2.70	2.63	3.44	2.10

Table A-15: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water with initial bacteriophage Qβ concentration at 8 x 10^7 PFU/mL and filtrated water by 1.0 μm ceramic membrane microfiltration at various PACl concentration.

Parameters	PACl dosage (mg-Al/L)				
	0.0	**1.5**	**2.0**	**2.5**	**3.0**
Turbidity (NTU)	0.81	1.72	1.00	0.70	0.79
Temperature (°C)	24.7	19.2	21.0	21.5	21.2
pH	7.70	7.67	7.6	7.5	7.42
EC (μs/cm)	248	269	246	252	245
DOC (mg/L)	2.317	1.645	1.474	1.218	1.205
Alkalinity (mg/L as CaCO3)	110.4	124.2	124.2	124.2	124.2
UV-254(cm^{-1})	0.066	0.043	0.034	0.032	0.034
SUVA (L/mg.m)	2.84	2.61	2.30	2.62	2.82

Table A-16: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water with initial bacteriophage Qβ concentration at 8 x 10^7 PFU/mL and filtrated water by 0.5 μm ceramic membrane microfiltration at various PACl concentration.

Parameters	PACl dosage (mg-Al/L)				
	0.0	**1.5**	**2.0**	**2.5**	**3.0**
Turbidity (NTU)	0.19	0.04	<0.01	0.01	0.15
Temperature (°C)	23.9	26.7	26.4	26.5	20.6
pH	7.69	7.49	7.38	7.38	7.25
EC (μs/cm)	244	237	241	220	274
DOC (mg/L)	2.267	1.637	1.591	1.239	1.114
Alkalinity (mg/L as CaCO3)	119.0	124.2	119.6	115	138
UV-254(cm^{-1})	0.064	0.044	0.043	0.038	0.036
SUVA (L/mg.m)	2.82	2.68	2.70	3.06	3.23

Table A-17: DOC, UV-254, SUVA, Turbidity, EC, Alkalinity, Temperature and pH of spiked-surface water with initial bacteriophage Qβ concentration at 8 x 10^7 PFU/mL and filtrated water by 0.1 μm ceramic membrane microfiltration at various PACl concentration.

Parameters	PACl dosage (mg-Al/L)				
	0.0	**1.5**	**2.0**	**2.5**	**3.0**
Turbidity (NTU)	0.07	0.07	0.09	<0.01	<0.01
Temperature (°C)	24.5	19.8	20.4	20.4	20.6
pH	7.68	7.39	7.37	7.44	7.14
EC (μs/cm)	245	256	240	250	253
DOC (mg/L)	2.256	1.524	1.347	1.179	1.089
Alkalinity (mg/L as CaCO3)	115.0	149.5	126.5	149.5	138
UV-254(cm^{-1})	0.060	0.036	0.034	0.031	0.037
SUVA (L/mg.m)	2.67	2.362	2.52	2.62	3.39

APPENDIX B

FLUX AND COAGULTION DETENTION TIME OF CERAMIC MEMBRANE MICROFILTRATION

Table B-1: Flux and coagulation detention time of spiked-Qβ water ($5x10^5$ PFU/mL)**.**

Water samples	Flux (cm/s)	coagulation detention time (s)
Raw water		
Raw water 1.0 μm	59.39	29.88
1.5 mg-Al/L PACl + 1.0 μm	39.48	22.64
2.0 mg-Al/L PACl + 1.0 μm	41.09	20.94
2.5 mg-Al/L PACl + 1.0 μm	38.20	19.70
3.0 mg-Al/L PACl + 1.0 μm	41.09	20.15
Raw water		
Raw water 0.5 μm	23.72	33.73
1.5 mg-Al/L PACl + 0.5 μm	31.41	25.47
2.0 mg-Al/L PACl + 0.5 μm	33.98	23.55
2.5 mg-Al/L PACl + 0.5 μm	33.98	23.55
3.0 mg-Al/L PACl + 0.5 μm	28.38	28.19
Raw water		
Raw water 0.1 μm	26.77	13.47
1.5 mg-Al/L PACl + 0.1 μm	41.82	20.26
2.0 mg-Al/L PACl + 0.1 μm	45.30	19.47
2.5 mg-Al/L PACl + 0.1 μm	43.90	20.54
3.0 mg-Al/L PACl + 0.1 μm	43.62	19.47

Table B-2: Flux and coagulation detention time of spiked-Qβ water $4x10^6$ PFU/mL).

Water samples	Flux (cm/s)	coagulation detention time (s)
Raw water		
Raw water 1.0 μm	59.39	20.13
1.5 mg-Al/L PACl + 1.0 μm	38.62	22.64
2.0 mg-Al/L PACl + 1.0 μm	40.38	20.94
2.5 mg-Al/L PACl + 1.0 μm	45.01	19.74
3.0 mg-Al/L PACl + 1.0 μm	40.62	20.15
Raw water		
Raw water 0.5 μm	23.72	33.73
1.5 mg-Al/L PACl + 0.5 μm	38.62	20.72
2.0 mg-Al/L PACl + 0.5 μm	40.38	19.81
2.5 mg-Al/L PACl + 0.5 μm	45.01	17.77
3.0 mg-Al/L PACl + 0.5 μm	40.62	19.70
Raw water		
Raw water 0.1 μm	26.77	13.47
1.5 mg-Al/L PACl + 0.1 μm	37.79	21.17
2.0 mg-Al/L PACl + 0.1 μm	38.62	20.72
2.5 mg-Al/L PACl + 0.1 μm	37.59	21.28
3.0 mg-Al/L PACl + 0.1 μm	39.93	20.04

Table B-3: Flux and coagulation detention time of spiked-Qβ water ($8x10^6$ PFU/mL).

Water samples	Flux (cm/s)	coagulation detention time (s)
Raw water		
Raw water 1.0 μm	59.39	29.24
1.5 mg-Al/L PACl + 1.0 μm	43.09	18.56
2.0 mg-Al/L PACl + 1.0 μm	48.08	16.64
2.5 mg-Al/L PACl + 1.0 μm	38.00	21.06
3.0 mg-Al/L PACl + 1.0 μm	49.42	16.19
Raw water		
Raw water 0.5 μm	23.72	34.36
1.5 mg-Al/L PACl + 0.5 μm	34.81	22.98
2.0 mg-Al/L PACl + 0.5 μm	39.04	20.49
2.5 mg-Al/L PACl + 0.5 μm	47.43	16.87
3.0 mg-Al/L PACl + 0.5 μm	40.38	19.81
Raw water		
Raw water 0.1 μm	26.77	13.47
1.5 mg-Al/L PACl + 0.1 μm	41.82	19.13
2.0 mg-Al/L PACl + 0.1 μm	45.30	17.66
2.5 mg-Al/L PACl + 0.1 μm	43.90	18.23
3.0 mg-Al/L PACl + 0.1 μm	43.62	18.34

Table B-4: Flux and coagulation detention time of spiked-Qβ water ($8.0x10^7$ PFU/mL).

Water samples	Flux (cm/s)	coagulation detention time (s)
Raw water		
Raw water 1.0 μm	59.39	22.13
1.5 mg-Al/L PACl + 1.0 μm	38.62	21.64
2.0 mg-Al/L PACl + 1.0 μm	40.38	21.94
2.5 mg-Al/L PACl + 1.0 μm	45.01	19.94
3.0 mg-Al/L PACl + 1.0 μm	40.62	20.45
Raw water		
Raw water 0.5 μm	23.72	34.23
1.5 mg-Al/L PACl + 0.5 μm	38.62	21.32
2.0 mg-Al/L PACl + 0.5 μm	40.38	20.21
2.5 mg-Al/L PACl + 0.5 μm	45.01	18.63
3.0 mg-Al/L PACl + 0.5 μm	40.62	20.50
Raw water		
Raw water 0.1 μm	26.77	16.47
1.5 mg-Al/L PACl + 0.1 μm	37.79	20.86
2.0 mg-Al/L PACl + 0.1 μm	38.62	21.56
2.5 mg-Al/L PACl + 0.1 μm	37.59	20.64
3.0 mg-Al/L PACl + 0.1 μm	39.93	21.44

APPENDIX C

BACTERIOPHAGE Qβ CONCENTRATION AND LOG BACTERIOPHAGE Qβ REMOVAL OF CERAMIC MEMBRANE MICROFILTRATION

Table C-1: Reduction of the bacteriophage Qβ from the initial bacteriophage Qβ concentration 8.0x 10^6 PFU/mL in raw water.

Water Samples	Bacteriophage Qβ (PFI/mL)	Log removal
Raw water	$8.00x10^6$	
Raw water 1.0 μm	$1.78x10^6$	0.7
1.5 mg-Al/L PACl + 1.0 μm	$1.23x10^3$	3.8
2.0 mg-Al/L PACl + 1.0 μm	$2.56x10^2$	4.5
2.5 mg-Al/L PACl + 1.0 μm	$1.20x10^1$	5.8
3.0 mg-Al/L PACl + 1.0 μm	$1.00x10^0$	6.9
Raw water	$8.0x\ 10^6$	
Raw water 0.5 μm	$1.18x10^6$	0.8
1.5 mg-Al/L PACl + 0.5 μm	$1.16x10^2$	4.8
2.0 mg-Al/L PACl + 0.5 μm	$1.60x10^1$	5.7
2.5 mg-Al/L PACl + 0.5 μm	$1.00x10^0$	6.9
3.0 mg-Al/L PACl + 0.5 μm	$1.00x10^0$	6.9
Raw water	$8.00x10^6$	
Raw water 0.1 μm	$9.33x10^5$	0.9
1.5 mg-Al/L PACl + 0.1 μm	$1.60x10^1$	5.7
2.0 mg-Al/L PACl + 0.1 μm	$1.00x10^0$	6.9
2.5 mg-Al/L PACl + 0.1 μm	$1.00x10^0$	6.9
3.0 mg-Al/L PACl + 0.1 μm	$1.00x10^0$	6.9

Table C-2: Reduction of the bacteriophage Qβ from the initial bacteriophage Qβ concentration 4.0x 10^6 PFU/mL in raw water.

Water Samples	Bacteriophage Qβ (PFI/mL)	Log removal
Raw water	$4.00x10^6$	
Raw water 1.0 μm	$1.31x10^6$	0.5
1.5 mg-Al/L PACl + 1.0 μm	$2.60x10^2$	4.2
2.0 mg-Al/L PACl + 1.0 μm	$3.80x10^1$	5.0
2.5 mg-Al/L PACl + 1.0 μm	$5.00x10^0$	5.9
3.0 mg-Al/L PACl + 1.0 μm	$1.00x10^0$	6.6
Raw water	$4.00x10^6$	
Raw water 0.5 μm	$9.34x10^6$	0.6
1.5 mg-Al/L PACl + 0.5 μm	$2.57x10^5$	5.2
2.0 mg-Al/L PACl + 0.5 μm	$3.75x10^1$	6.0
2.5 mg-Al/L PACl + 0.5 μm	$1.00x10^0$	6.6
3.0 mg-Al/L PACl + 0.5 μm	$1.00x10^0$	6.6
Raw water	$4.00x10^6$	
Raw water 0.1 μm	$8.33x10^5$	0.7
1.5 mg-Al/L PACl + 0.1 μm	$4.45x10^3$	6.0
2.0 mg-Al/L PACl + 0.1 μm	$1.00x10^0$	6.6
2.5 mg-Al/L PACl + 0.1 μm	$1.00x10^0$	6.6
3.0 mg-Al/L PACl + 0.1 μm	$1.00x10^0$	6.6

Table C-3: Reduction of the bacteriophage Qβ from the initial bacteriophage Qβ concentration 5.0x 10^5 PFU/mL in raw water.

Water Samples	Bacteriophage Qβ (PFI/mL)	Log removal
Raw water	$5.00x10^6$	
Raw water 1.0 μm	$1.80x10^5$	0.4
1.5 mg-Al/L PACl + 1.0 μm	$7.80x10^1$	3.8
2.0 mg-Al/L PACl + 1.0 μm	$1.00x10^0$	5.7
2.5 mg-Al/L PACl + 1.0 μm	$1.00x10^0$	5.7
3.0 mg-Al/L PACl + 1.0 μm	$1.00x10^0$	5.7
Raw water	$5.00x\ 10^6$	
Raw water 0.5 μm	$1.57x10^5$	0.5
1.5 mg-Al/L PACl + 0.5 μm	$3.00x10^1$	4.1
2.0 mg-Al/L PACl + 0.5 μm	$1.00x10^0$	5.7
2.5 mg-Al/L PACl + 0.5 μm	$1.00x10^0$	5.7
3.0 mg-Al/L PACl + 0.5 μm	$1.00x10^0$	5.7
Raw water	$5.00x10^6$	
Raw water 0.1 μm	$1.38x10^5$	0.6
1.5 mg-Al/L PACl + 0.1 μm	$1.00x10^0$	5.7
2.0 mg-Al/L PACl + 0.1 μm	$1.00x10^0$	5.7
2.5 mg-Al/L PACl + 0.1 μm	$1.00x10^0$	5.7
3.0 mg-Al/L PACl + 0.1 μm	$1.00x10^0$	5.7

Table C-4: Reduction of the bacteriophage Qβ from the initial bacteriophage Qβ concentration 8.0x 10^5 PFU/mL in raw water.

Water Samples	Bacteriophage Qβ (PFI/mL)	Log removal
Raw water	$8.00x10^7$	
Raw water 1.0 µm	$6.07x10^7$	0.1
1.5 mg-Al/L PACl + 1.0 µm	$3.45x10^4$	3.4
2.0 mg-Al/L PACl + 1.0 µm	$1.65x10^3$	4.7
2.5 mg-Al/L PACl + 1.0 µm	$4.86x10^2$	5.2
3.0 mg-Al/L PACl + 1.0 µm	$7.60x10^1$	6.0
Raw water	$8.00x10^7$	
Raw water 0.5 µm	$5.53x10^7$	0.2
1.5 mg-Al/L PACl + 0.5 µm	$5.55x10^3$	4.2
2.0 mg-Al/L PACl + 0.5 µm	$4.37x10^2$	5.3
2.5 mg-Al/L PACl + 0.5 µm	$5.30x10^1$	6.2
3.0 mg-Al/L PACl + 0.5 µm	$1.00x10^0$	7.9
Raw water	$8.00x10^7$	
Raw water 0.1 µm	$5.06x10^7$	0.2
1.5 mg-Al/L PACl + 0.1 µm	$1.88x10^3$	4.6
2.0 mg-Al/L PACl + 0.1 µm	$2.42x10^2$	5.5
2.5 mg-Al/L PACl + 0.1 µm	$3.50x10^1$	6.4
3.0 mg-Al/L PACl + 0.1 µm	$1.00x10^0$	7.9

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Presentation:

Paveetida Yanthongyu and Suraphong Wattanachira, BACTERIOPHAGE REMOVAL EFFICIENCY BY IN-LINE PACI COAGULATIONWITH CERAMIC MEMBRANE FILTRATION. The proceeding of International Conference on Green and Sustainable Innovation 2012 in Chiang Mai, Thailand, May 24 –26, 2012