การอุดตันซิลิกาของเมมเบรนออสโมซิสย้อนกลับชนิดความดันต่ำ UTC-70 ในการบำบัดฟลูออไรด์ในน้ำใต้ดินแอ่งเชียงใหม่

นายภาคภูมิ รักร่วม

วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิทยาศาสตรมหาบัณฑิต
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SILICA FOULING OF UTC-70 ULTRA LOW PRESSURE REVERSE OSMOSIS MEMBRANE IN DEFLUORIDATION OF CHIANG MAI BASIN GROUNDWATER

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ภาคภูมิ รักร่วม : การอุดตันซิลิกาของเมมเบรนออสโมซิสย้อนกลับชนิดความดันต่ำ UTC-70 ในการบำบัดฟลูออไรด์ในน้ำใต้ดินแอ่งเชียงใหม่. (SILICA FOULING OF UTC-70 ULTRA LOW PRESSURE REVERSE OSMOSIS MEMBRANE IN DEFLUORIDATION OF CHIANG MAI BASIN GROUNDWATER) อ.ที่ปรึกษาวิทยานิพนธ์หลัก: รศ.ดร.สุรพงษ์ วัฒนะจีระ, 115 หน้า.

เมมเบรนออสโมซิสย้อนกลับชนิดแรงดันต่ำเป็นเทคโนโลยีใหม่ที่ถูกนำมาประยุกต์ใช้ในการบำบัดฟลูออไรด์ในน้ำใต้ดิน โดยมีข้อดีกว่าการใช้เมมเบรนออสโมซิสคือสามารถเดินระบบได้ที่ความดันที่ต่ำคือ 0.1-0.5 เมกะปาสคาล แต่ปัญหาที่ยังพบอยู่ในการเดินระบบก็คือ การอุดตันที่ผิวของเมมเบรน ซึ่งมีผลกระทบต่ออัตราการผลิตน้ำสะอาด รวมไปถึงการทำให้เมมเบรนเกิดการเสียหาย ในงานวิจัยนี้จึงได้ทำการศึกษาถึงการอุดตันของเมมเบรนออสโมซิสย้อนกลับชนิดแรงดันต่ำในการบำบัดฟลูออไรด์ในน้ำใต้ดิน โดยได้เลือกตัวอย่างน้ำใต้ดินสองจุดในแอ่งเชียงใหม่มาเป็นน้ำตัวอย่าง เนื่องจากน้ำใต้ดินบริเวณแอ่งเชียงใหม่มีปริมาณฟลูออไรด์อยู่ในระดับที่สูง น้ำใต้ดินจากทั้งสองแหล่งจะถูกนำมากรองผ่านเมมเบรนออสโมซิสย้อนกลับชนิดแรงดันต่ำ โดยเดินระบบแบบครอสโฟลว์ และแปรผันค่าความดัน 2 ค่าคือ 0.3 และ 0.5 เมกะปาสคาล นอกจากนั้นยังมีการศึกษาถึงปรากฏการณ์คอนเซนเตรชั่น โพลาไรเซชั่น โดยใช้น้ำที่มีค่าความเข้มข้นของฟลูออไรด์ต่างๆกัน คือ 0, 10, 25, และ 50 มิลลิโมล เป็นน้ำตัวอย่าง จากการทดลองจะพบว่าค่าสัมประสิทธิ์การเคลื่อนย้ายสารฟลูออไรด์ที่ความดัน 0.3 และ 0.5 เมกะปาสคาล มีค่า 2.73×10^{-5} และ 1.75×10^{-5} เมตรต่อวินาทีตามลำดับ ส่วนค่าเฉลี่ยของสัมประสิทธิ์การเคลื่อนย้ายฟลูออไรด์ผ่านเมมเบรนมีค่า 4.5×10^{-7} เมตรต่อวินาที ตามลำดับ สำหรับในการศึกษาการบำบัดฟลูออไรด์ในน้ำใต้ดิน จะพบว่าได้ค่าอัตราการบำบัดสารฟลูออไรด์ที่สูงมากกว่า 90% โดยอัตราการบำบัดจะมีค่าสูงที่ความดันสูง (0.5 เมกะปาสคาล) นอกจากนั้นยังมีการศึกษาการอุดตันที่ผิวของเมมเบรน โดยจะพบว่าการอุดตันที่เกิดขึ้นเป็นแบบแน่น (polymerized fouled layer) ในทุกการทดลอง จากการศึกษาผลของการอุดตันซิลิกาในการบำบัดฟลูออไรด์ในน้ำใต้ดิน จะพบว่าปริมาณซิลิกาที่อยู่ในน้ำเข้าที่เข้าระบบ จะมีผลต่ออัตราการบำบัดฟลูออไรด์ โดยปริมาณซิลิกาที่เพิ่มขึ้นจะมีผลทำให้อัตราการบำบัดฟลูออไรด์ลดต่ำลง และยังพบอีกว่าการอุดตันของซิลิกาบริเวณผิวเมมเบรนเป็นการอุดตันแบบแน่น (polymerized silica fouled layer)

สาขาวิชา การจัดการสิ่งแวดล้อม ลายมือชื่อนิสิต ภาคภูมิ รักร่วม
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KEYWORDS : CONCENTRATION POLARIZATION / FLUORIDE REJECTION RATE / GROUNDWATER DEFLUORIDATION / SILICA FOULING / ULPRO

PHARKPHUM RAKRUAM: SILICA FOULING OF UTC-70 ULTRA LOW PRESSURE REVERSE OSMOSIS MEMBRANE IN DEFLUORIDATION OF CHIANG MAI BASIN GROUNDWATER. ADVISOR: ASSOC.PROF.SURAPHONG WATTANACHIRA, 115 PP.

Ultra low pressure reverse osmosis membrane is a new technology, which apply to defluoridation from groundwater. This membrane can operated at very low pressure. However, the major problem of reverse osmosis is membrane fouling. Membrane fouling can effect on fluoride rejection rate and cause of membrane degradation. In this study, the membrane fouling of UTC-70 ultra low pressure reverse osmosis membrane in groundwater defluoridation was investigated. Chiang mai basin was selected as study area. In this area, groundwater contains high fluoride concentration. Two sites of groundwater in this area were selected as sampling point. Fisrt site is groundwater from Pra Too Khong, Lamphun, wheareas, the second site is San Kam Pang hot spring well, Chiang mai. Groundwater from two sites was filtrated with ULPRO membrane, which operated in cross-flow mode. The operating transmembrane pressure was varied at 0.3 and 0.5 MPa. Furthermore, the concentration polarization phenomenon was investigated. The NaF concentration varied at 0, 10, 25, and 50 mM was used as feed solution. From the results, it was found that the mass transfer coefficient in solution was 2.73×10^{-5} and 1.75×10^{-5} m/s at 0.3 and 0.5 MPa, respectively. While, the average solute mass transfer coefficient through the membrane was 4.5×10^{-7} m/s under 0.3 and 0.5 MPa, respectively. For the groundwater defluoridation experiment, it was found that the high fluoride rejection rate more than 90% was obtained. In Pra Too Khong groundwater, the fluoride rejection rate was 95.7-96.7%, wheareas, in San Kam Pang hot spring well was 96.7-97.5%. From the results, it can be indicated that the higher fluoride rejection rate was obtained at higher transmembrane pressure (0.5 MPa). From the investigation of membrane fouling, it was found that the membrane was fouled with dense-gel layer (polymerized fouled layer) in all case of experiment. Furthermore, the effect of silica fouling on groundwater defluoridation was investigated. The results showed that silica can effect on permeate flux and fluoride rejection rate in groundwater defluoridation. The presence of silica in feed water was cause of decreasing in fluoride rejection rate. While, the results of silica fouling showed that membrane was fouled with polymerized silica fouled layer.

Field of Study : Environmental Management Student's Signature : Pharkphum
Academic Year : 2008 Advisor's Signature:

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CONTENTS

Page

Page

LIST OF TABLES

Table page

LIST OF FIGURES

Figure Page

NOMENCLATURES

μg/g	Microgram per Gram
μm	Micro meter
˚C	Degree Celsius
ΔP	Transmembrane pressure
π	Osmotic pressure
π_B	Osmotic pressure of bulk solution
π_P	Osmotic pressure of permeate water
f	Concentration polarization level
Alk	Alkalinity
$CaCO_3$	Calcium carbonate
CaF_2	Calcium fluoride
$Ca_5\,(PO_4).F$	Apatite
C_B	Fluoride concentration on concentrated water
C_{Gel}	Fluoride concentration on gel layer surface
C_M	Fluoride concentration on membrane surface
C_P	Fluoride concentration in permeate water
cm^2	Square centimeter
DOC	Dissolve organic carbon
EC	Electro conductivity
EHC	Environment Health Criteria
Fe (II)	Iron
Ji	Solute flux
Jv	Permeate flux
$(Jv)_{H20}$	Permeate flux of pure water
k	Mass transfer coefficient
ki	Solute mass transfer coefficient
km^2	Square kilometer
MF	Micro filtration

m^3/day	Cubic meter per day
mg/day	Milligram per day
mg/L	Milligram per Liter
m/s	Meter per seconds
mM	Milli molar
ml	Milli liter
MPa	Mega Pascal
mv	Milli volt
Na_3AlF_6	Cryolite
NaF	Sodium fluoride
NF	Nano filtration
NH4-N	Ammonium
Nm	Nano meter
ORP	Oxidation reduction potential
Rint	Intrinsic rejection rate
Robs	Observed rejection rate
RO	Reverse Osmosis
SiO_2	Silicon dioxide
UF	Ultra filtration
ULPRO	Ultra Low Pressure Reverse Osmosis
UTC	Ultra Thin Composite
WHO	World Health Organization

CHAPTER I

INTRODUCTION

1.1 Motivation

Fluoride is a common element that does not occur in the elemental state in nature because of its high reactivity (WHO, 2004). Then, fluoride is rarely found in pure form, but exits in various compounds or as the monovalent anion fluoride. It is found in Earth's crust in form of fluorspar (CaF_2), cryolite (Na_3AlF_6), and fluorapatite ($Ca_5 (PO_4).F$).

Fluoride is naturally released into environment through the weathering and dissolution of minerals. It is also released via coal combustion and waste from industrial processes; for example the effluent of steel manufacture and primary aluminium production. Fluoride levels in surface waters vary according to geographical location and proximity to emission sources (EHC 227, 2002), whereas in groundwater, fluoride concentrations vary with the type of rock the water flows through. In Chiang mai basin, fluorides concentration in groundwater was high to 16.1 mg/L. Groundwater in some districts of Lamphun province, which located in Chiang mai basins, is found to have fluoride levels of between 10 to 15 mg/L, which is higher than the drinking water limit of the World Health Organization (1.5 mg/L). So, villagers in this area were affected from the dental and skeletal fluorosis. The groundwater in this area is recommended to defluoride before drinking.

Membrane filtration with reverse osmosis membrane was applied to defluoridation in Lamphun province. The quality of filtrated water was high with less amount of fluoride in water. However, the process of reverse osmosis is required high pressure (1.5-15 MPa) to operating. So, the operating costs are high and make the villagers have difficulty to access this technology. From the problem of reverse osmosis membrane technology, the ultra low pressure reverse osmosis (ULPRO) membrane is applied to defluoridation. The ULPRO membrane can be operated at very low pressure (0.1-0.5 MPa) and the high fluoride rejection rates are obtained.

However, the major problem in membrane filtration is membrane fouling. This problem can reduce the permeate flux, permeate quality and membrane degradation. Membrane fouling is occurred by many ions such as calcium, magnesium and silica. Most of membrane plant in Lamphun province is contained the pretreatment process before enter to reverse osmosis system. The pretreatment process can be removing some ions including sodium, calcium, magnesium and potassium but can not remove silica. Then, silica fouling has been the major problem in groundwater defluoridation system.

Silica is widely found in the environment. In natural water sources, silica is released from the weathering of minerals, which containing silica species (E.G. Darton, 1999). Silica is one of the foulants present in the feed water in RO membrane plants. Silica fouling can reduce permeate flux in membrane filtration processes. Then, the objectives of this research are to investigate the membrane fouling of ultra low pressure reverse osmosis membrane in groundwater defluoridation, especially in the effect of silica fouling. Groundwater from Pra Too Khong drinking water plant and groundwater from San Kam Pang hot spring well, which defined as high silica concentration, was selected as sampling point.

1.2 Objectives

- To study the performance of UTC-70 ultra low pressure reverse osmosis membrane in groundwater defluoridation.
- To investigate the membrane fouling and effect of silica on groundwater defluoridation by ultra low pressure reverse osmosis membrane.
- To determine the mass transfer coefficient (k) in solution and solute mass transfer coefficient through membrane (ki).

1.3 Hypothesis

- The ULPRO membrane is a new technogy, which can be operated at low pressure (0.1-0.5 MPa) and provided the high fluoride rejection rate.
- Silica can be effect on performance of groundwater defluoridation by forming the silica gel layer on membrane surface, which lead to decreased in permeate flux or fluoride rejection rates in groundwater defluoridation.
- Concentration polarization phenomenon may occur during the process and effect on groundwater defluoridation.

1.4 Scope of the study

The study area of this research is Chiang Mai basin. The Chiang Mai Basin is located in northern Thailand, which covers about 3,000 km^2 of Chiang Mai and Lamphun province (Matsui, 2007). Two sites of groundwater in Chiang Mai basin was selected as sampling point. The first site is groundwater from Pra Too Khong Bottled Drinking Water Plant, Tambon Ban Klang, Muang district, Lamphun province (site 1). The second site is groundwater from San Kam Pang hot spring well (site 2). The results from 2 sites were compared.

The experiment was conducted in a cross-flow unit with a flat sheet of UTC-70 membrane, which is ultra low pressure reverse osmosis membrane. The transmembrane pressure was varied at 0.3 and 0.5 MPa. The performance of groundwater defluoridation by ultra low pressure reverse osmosis membrane was investigated. Furthermore, the effect of silica on groundwater defluoridation was investigated by added silica concentration 500 mg/L into groundwater from both sites.

The concentration polarization phenomenon was studied. The NaF concentration varied at 0, 10, 25, 50 mM was used as feed solution.

CHAPTER 2

BACKGROUND AND LITERATURE REVIEWS

2.1 Fluoride

2.1.1 Background

Fluoride is hazardous inorganic specie that seriously endangers the aquatic environment. Fluoride is found in Earth'crust and exists in the most common form of fluorspar, cryolite and fluorapatite. Fluoride is found naturally in rocks, seawater, surface water, and groundwater (Ndiaye et al., 2005).

Fluorides are released into the environment naturally through the weathering and dissolution of minerals, in emissions from volcanoes and in marine aerosols (Symonds et al., 1988; ATSDR, 1993). Fluorides are also released into the environment via coal combustion and process waters and waste from various industrial processes, including steel manufacture, primary aluminium, copper and nickel production, phosphate ore processing, phosphate fertilizer production and use, glass, brick and ceramic manufacturing, and glue and adhesive production (EHC 227, 2002).

Fluoride was used in many processes in some industrial: Hydrogen fluoride is an important industrial compound that is used mainly in the motor gasoline alkylates and chlorofluorocarbons. Calcium fluoride is used as a flux in steel, glass and enamel production, as the raw material for the production of an electrolyte in aluminium production. Sodium fluoride is used in the controlled fluoridation of drinking-water, as a preservative in glues, in glass and enamel production, as a flux in steel and aluminium production, as an insecticide and as a wood preservative. Sulfur hexafluoride is used extensively in various electronic components and in the production of magnesium and aluminium. Fluorosilicic acid (H_2SiF_6) and sodium hexafluorosilicate (Na_2SiF_6) are used for the fluoridation of drinking-water supplies

Fluoride levels in surface waters vary according to location and proximity to emission sources (EHC 227, 2002). Fluoride content in the bedrock was the most important factor determining fluoride levels in lake waters. Elevated fluoride concentrations were found in acidified areas compared with other regions with similar geology (Skjelkvåle, 1994a). Elevated inorganic fluoride levels in surface water are often seen in regions where there is geothermal or volcanic activity, at the foot of high mountains and in areas with geological deposits of marine origin. Surface water concentrations generally range from 0.01 to 0.3 mg/litre. Seawater contains more fluoride than fresh water, with concentrations ranging from 1.2 to 1.5 mg/litre. Higher levels of fluoride are found in areas where the natural rock is rich in fluoride.

World Health Organization set the guideline value of fluoride concentration at 1.5 mg/L (WHO, 2004). Concentrations above this value are risk of dental and skeletal fluorosis. Due to Thailand are located in tropical zone, the Ministry of Public health has set as 0.7 mg/L for fluoride concentration in drinking water.

2.1.2 Cycle in environment

Figure 2.1 showed the cycle of fluoride in environment and it can be indicate that all components have a relationship with each of other components.

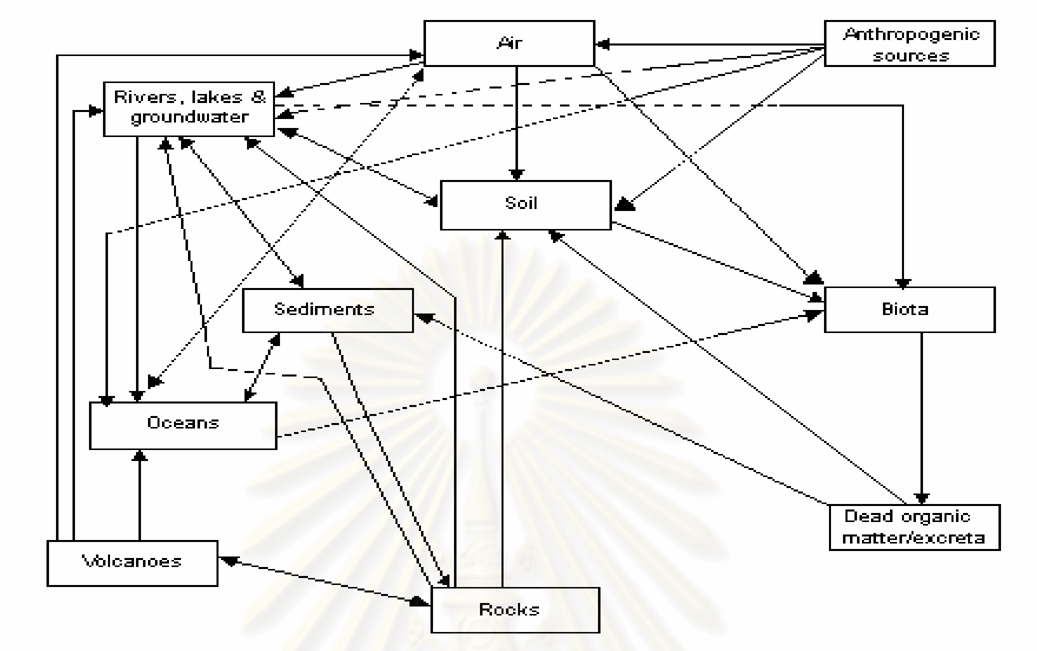


Figure 2.1 The cycle of fluoride in environment

Fluorides in the atmosphere may be in gaseous or particulate form. Gaseous forms include hydrogen fluoride, silicon tetrafluoride (SiF_4), fluorosilicic acid and sulfur hexafluoride. Particulate forms include sodium aluminium fluoride (cryolite), aluminium fluoride, calcium fluoride, sodium hexafluorosilicate, lead fluoride (PbF_2) and calcium phosphate fluoride (fluorapatite). Atmospheric fluorides can be transported over large distances as a result of wind or atmospheric turbulence or can be removed from the atmosphere via wet and dry deposition or hydrolysis. The fate of inorganic fluorides in the atmosphere is primarily influenced by vaporization, aerosol formation, wet and dry deposition and hydrolysis (Environment Canada, 1994). Non-volatile inorganic fluoride particulates are removed from the atmosphere via condensation or nucleation processes. Atmospheric fluorides may be transported to soils and surface waters through both wet and dry deposition processes (US NAS, 1971).

In water, the transport and transformation of inorganic fluorides are influenced by pH, water hardness and the presence of ion-exchange materials such as clays (Environment Canada, 1994). Fluoride is usually transported through the water cycle complexed with aluminium (Ares, 1990). Once dissolved, inorganic fluorides remain in solution under conditions of low pH and hardness and in the presence of ion-exchange material (Cuker & Shilts, 1979). Fluoride levels in surface waters vary according to geographical location and proximity to emission sources. Seawater contains more fluoride than fresh water, with concentrations ranging from 1.2 to 1.5 mg/litre. In seawater, fluorides are removed by the formation of complexes with calcium compounds, principally carbonate and phosphate (Carpenter, 1969). Undissolved fluoride is generally removed from the aquatic phase by sedimentation (US EPA, 1980).

Factors that influence the mobility of inorganic fluorides in soil are pH and the formation of aluminium and calcium complexes (Pickering, 1985; Environment Canada, 1994). Fluoride in soil is mainly bound in complexes. The maximum adsorption of fluoride to soil was reported to occur at pH 5.5 (Barrow & Ellis, 1986). Fluoride is a component of most types of soil, with total concentrations ranging from 20 to 1000 µg/g in areas without natural phosphate or fluoride deposits and up to several thousand micrograms per gram in mineral soils with deposits of fluoride (Davison, 1983).

2.1.3 Human exposure

Levels of daily exposure to fluoride depend mainly on the geographical area. Fluorides can be taken up by aquatic organisms directly from the water or to a lesser extent via food. In animals, fluorides can be accumulating in the bone tissue. The principal route for the intake of fluoride is the consumption of foodstuffs and drinking-water. In areas of the world in which coal rich in fluoride is used for heating and food preparation, the inhalation of indoor air and consumption of foodstuffs containing increased levels of fluoride also contribute to elevated intakes. In general, estimated intakes of fluoride in children and adolescents do not exceed approximately 2 mg/day.

2.1.3.1 Drinking water

Fluoride is present in all phase of the environment. Therefore, sources of drinking-water are likely to contain at least some small amount of fluoride. The amount of fluoride present naturally in non-controlled fluoridated drinking-water (i.e., drinking-water to which fluoride has not been intentionally added for the prevention of dental caries) is highly variable, being dependent upon the individual geological environment from which the water is obtained (EHC 227, 2002). Table 2.1 showed the fluoride concentration in drinking water from some country.

Table 2.1 Fluoride concentration in drinking water from some country

Location	Fluoride concentration (mg/L)	References
USA	0.1-1.0	US EPA (1985); US DHHS (1991)
Canada	0.73-1.25	HealthCanada (1993)
Germany	0.02-0.17	Bergmann (1995)
Poland	0.02-3.0	Czarnowski et al. (1996)

2.1.3.2 Food products

Trace amount of fluoride are found in many foodstuffs. Levels of fluoride in foods are significantly affected by the fluoride content of the water used in preparation or processing. A study of a variety of infant food products available in the USA indicated concentrations of fluoride ranging from 0.01 to 8.38 µg/g; the highest concentrations were in products containing chicken (Heilman et al., 1997). The varius study of fluoride concentration in food are presented in Table 2.2

Table 2.2 Fluoride in food from some country

Food	Fluoride concentration (mg/L)	Remark	References
Milk and milk products	0.01-0.8	Range of concentrations in 12 varieties of dairy products in Canada	Dabeka & McKenzie (1995)
Fish	0.21-4.57	Range of concentrations in 4 varieties of fish available in Canada	Dabeka & McKenzie (1995)
Vegetables	0.28-1.34	Range of mean concentrations in three staple vegetables consumed in three villages in China	Chen et al. (1996)
Tea	0.37-2.07	Range of concentrations in black tea sampled between 1984 and 1989 in Germany	Bergmann (1995)
Beverages	0.21-0.96	Range of concentrations in 6 varieties of beer, wines, coffee and soft drinks available in Canada	Dabeka & McKenzie (1995)
Baked good and cereals	1.27-1.85	Range of mean concentrations in rice consumed in three villages in China	Chen et al. (1996)
Fruits and fruit juice	0.01–0.58	Range of concentrations in 25 varieties of fruit and fruit juices available in Canada	Dabeka & McKenzie (1995)

(Source: EHC 227, 2002)

2.1.3.3 Consumer products

Dental products such as toothpaste, mouthwash and fluoride supplements have been identified as significant sources of fluoride (Ekstrand, 1987;

Drummond et al., 1990). Topical mouth rinses marketed for daily home use may contain between 230 and 500 mg fluoride/litre, while mouthwash products intended for weekly or biweekly use may contain 900–1000 mg fluoride/litre (Sloof et al., 1989). For dental purposes, fluoride preparations may contain low (0.25–1 mg per tablet; 1000–1500 mg of fluorine per kg of toothpaste).

2.1.4 Health effects

The ingestion of excess fluoride can cause of various health effect include dental and skeletal fluorosis. The effect of fluoride in various concentrations on human health are showed in Table 2.3

Table 2.3 Health effect from various fluoride concentrations

Concentration of fluoride (mg/L)	Health effects
Low	Protection against dental caries
0.9-1.2	An adverse effect on tooth enamel and give rise to mild dental fluorosis
3-6	Skeletal fluorosis with adverse changes in bone structure
Over 10	Crippling skeletal fluorosis

(Source: International Program on Chemical Safety, 2002)

2.1.4.1 Dental fluorosis

Dental fluorosis occurs because of the excessive intake of fluoride either through fluoride in the water supply or through other sources during the formation of the tooth, usually from birth to approximately 6–8 years of age.

Excessive fluoride can cause yellowing of teeth, white spots, and pitting or mottling of enamel.

In general, dental fluorosis does not occur in temperate areas at concentrations below 1.5–2 mg of fluoride per litre of drinking-water. Because of the greater amounts of water consumed in warmer areas, dental fluorosis can occur at lower concentrations in the drinking-water (US EPA, 1985a).

Over the past 30–40 years, there has been an increase in the prevalence of dental fluorosis among populations consuming either fluoridated or non-fluoridated drinking-water. The prevalence of dental fluorosis is also elevated in certain areas of the world where the intake of fluoride may be inordinately high, due in large part to the elevated fluoride content of the surrounding geological environment. In China, large numbers of people exhibit dental fluorosis (Liu, 1995).

The severity of this condition, generally characterized as ranging from very mild to severe, is related to the extent of fluoride exposure during the period of tooth development. Mild dental fluorosis is usually typified by the appearance of small white areas in the enamel wheareas in severe dental fluorosis are stained and pitted appearance on teeth.

Figure 2.2, 2.3, and 2.4 showed the severity of dental fluorosis; mild fluorosis, moderate fluorosis and severe fluorosis, respectively.

Figure 2.2 Dental fluorosis – mild fluorosis

Figure 2.3 Dental fluorosis – moderate fluorosis

Figure 2.4 Dental fluorosis – severe fluorosis

Aunnop, 2006 reported that 55.39 % of students in the high fluoride concentration area have moderate to severe dental fluorosis. In contrast, 93.94 % of students in low fluoride concentration area have none or mild dental**.**

2.1.4.2 Skeletal fluorosis

Skeletal fluorosis is a clinical condition that may arise following long-term exposure (both inhalation and ingestion) to a high fluoride concentration. Elevated fluoride intakes can also have more serious effects on skeletal tissues. Skeletal fluorosis (with adverse changes in bone structure) may be observed when drinking-water contains 3–6 mg of fluoride per litre. Crippling skeletal fluorosis usually develops only where drinking-water contains over 10 mg of fluoride per litre (IPCS, 1984).

The severity of the effects associated with skeletal fluorosis is related to the amount of fluoride accumulated into bone. The first two stages are preclinical- that is, the patient feels no symptoms but changes have taken place in the body. In the early clinical stage of skeletal fluorosis, symptoms include pains in the bones and joints; sensations of burning, pricking, and tingling in the limbs; muscle weakness; chronic fatigue; and gastrointestinal disorders and reduced appetite. During this phase, changes in the pelvis and spinal column can be detected on x-rays. In the second clinical stage, pains in the bones become constant and some of the ligaments begin to calcify.

In advanced skeletal fluorosis, called crippling skeletal fluorosis, the extremities become weak and moving the joints is difficult. The vertebrae partially fuse together, crippling the patient.

Figure 2.5 Skeletal fluorosis

2.2 Membrane filtration

2.2.1 Background

Membrane filtration is technology that applied to purify contaminated water with undesirable components by passing water through a membrane as a filter material (Fujita et al., 1994). Water can pass through the membrane, while other components are rejected.

The membrane can be categorized into four types, microfiltration membrane (MF membrane), ultrafiltration membrane (UF membrane), nanofiltration membrane (NF membrane), and reverse osmosis membrane (RO membrane), by considering differential pressure on both sides of each membrane and the pore size of each membrane (Metcalf and Eddy, 2003).

The required pressure to operate the NF membrane (0.5-3.5 MPa) and the RO membrane (1.5-15 MPa) is higher than the required pressure for the MF membrane (< 0.2 MPa) and the UF membrane (0.1-1.0 MPa) (Wagner, 2001).

Generally, the MF membrane, the UF membrane, and the NF membrane are followed the principle of pores while the RO membrane is operated under the molecular interaction and diffusion. The characteristics of RO membrane, NF membrane, UF membrane and MF membrane are shown in Table 2.4.

Table 2.4 The characteristics of RO membrane, NF membrane, UF membrane and MF membrane

	RO membrane	NF membrane	UF membrane	MF membrane
Structure	Asymmetrical	Asymmetrical	Asymmetrical	Asymmetrical Symmetrical
Pore size	< 0.002 µm	< 0.002 µm	0.2 - 0.02µm	4 – 0.02 µm
Rejection	HMWC, LMWC glucose, amino acids	HMWC, polyvalent neg. ions	Macro molecules, polysaccharides	Particles, clay
Membrane material	Cellulose acetate, Thin film composite	Cellulose acetate, Thin film composite	Polysulfone, Thin film composite	Ceramic, Polysulfone
Operating pressure	1.5-15 MPa	0.5-3.5 MPa	0.1-1 MPa	< 0.2 MPa
Method	Dead-end filtration Cross-flow filtration	Dead-end filtration Cross-flow filtration	Dead-end filtration Cross-flow filtration	Dead-end filtration Cross-flow filtration

Source: Jørgen Wagner, 2001)

The MF membrane and the UF membrane are considered when undesirable components are larger particles. Because the permeate flux of these two membranes are high while the differential pressures are low. When ions are considered for removing from water, NF membrane and RO membrane are employed. In contrast, while the permeate water flux of these two membranes are low, the differential pressures are high.

Figure 1 illustrates the operating range of each membrane to remove undesirable components. It can be concluded that the RO membrane has the widest operating range to remove almost all undesirable components in water while the MF membrane has the narrowest operating range to remove undesirable components and the MF membrane should be strongly recommended as a pretreatment unit.

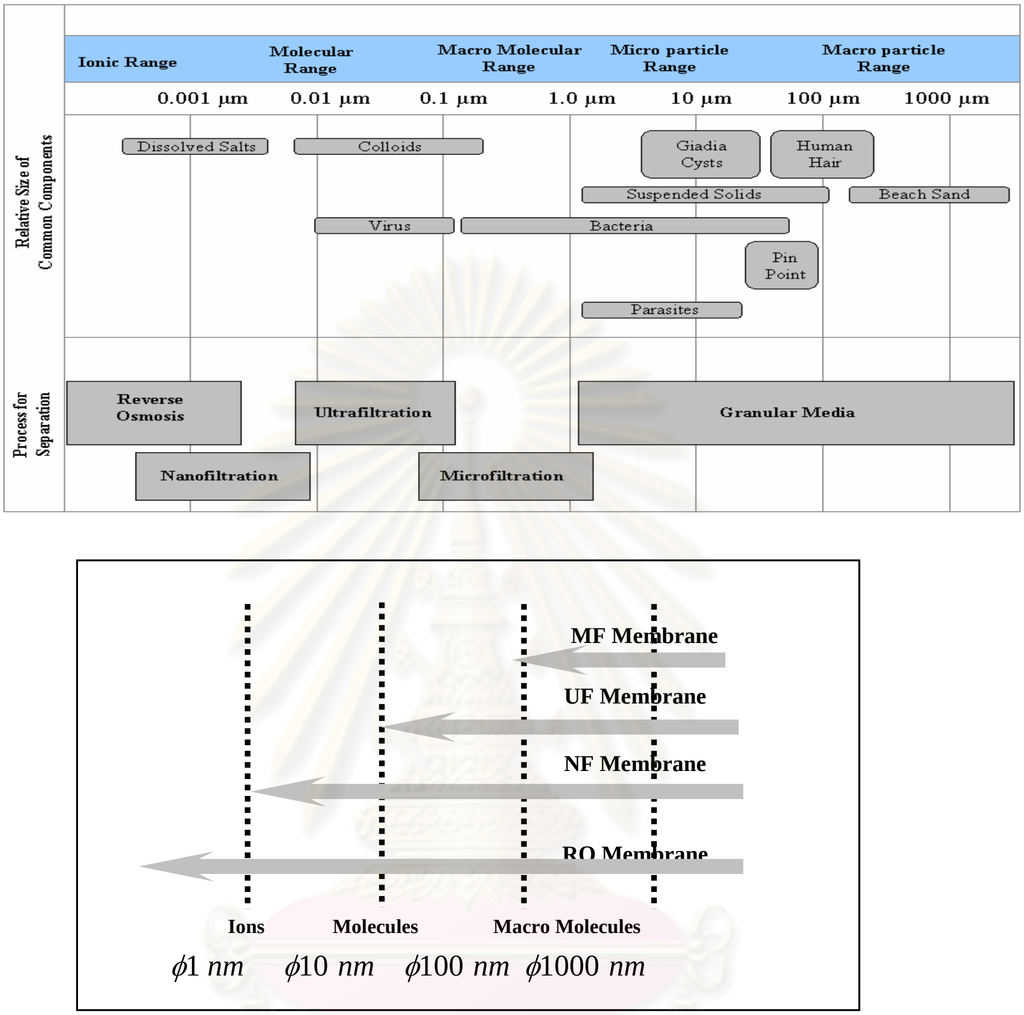


Figure 1. Operating ranges of membranes on undesirable components removal

2.2.2 Membrane transports theory

Many mathematical models have been used to describe membrane separation processes. Generally, when the membrane was used to separate solutes in solution, several forces including difference of pressure and difference of temperature can cause of a flow or flux of water and solutes through membrane.

2.2.2.1 Irreversible thermodynamic model

Irreversible thermodynamic model are considered the membrane separation processes, which in equilibrium or not far from the equilibrium. This model has been used to describe the transportation of solute through membrane. The model considers the membrane as a block box, then the membrane structure and membrane properties are not taken into account (Dickson, 1988; Williams, M. E., 2003).

Kedem and Katchalsky (1971) provided the equations for water flux based on membrane area (J_v) and the solute flux based on membrane area (J_i) in Equation (2-1) and Equation (2-2), respectively.

$$J_v = L_p(\mathit{\Delta}\mathrm{p} - \sigma \mathit{\Delta}\pi) \tag{2-1}$$

$$J_i = \omega \mathit{\Delta}\pi + (1-\sigma)\overline{C_i}J_v \tag{2-2}$$

Where L_p = pure water permeability, σ = reflection coefficient, ω = solute permeability, and $\overline{C_i}$ = logarithmic mean solute concentration. From the van't Hoff equation, $\mathit{\Delta}\pi = RT\mathit{\Delta}C_i$

2.2.2.2 Diffusive flow model

Generally, when component in solution is separated by membrane via diffusion mechanism, many parameters are involved including the driving forces of pressure, temperature, concentration, and electrical potential. The water flux (Jv) through the membrane can be expressed as

$$J_v = k_w\left(\Delta P - \Delta\pi\right) \tag{2-3}$$

where k_w = pure water permeability.
ΔP = transmembrane pressure
$\Delta\pi$ = osmotic pressure

And the solute flux (J_i) through the membrane, it can be expressed as

$$J_i = k_i \left(C_M - C_P \right) \tag{2-4}$$

where k_i = solute mass transfer coefficient.
C_M = concentration of membrane surface
C_P = concentration of permeate

2.2.2.3 Charged model

Charged membrane is characterized as a membrane that contains charged chemical groups. The removals of ionic species are mainly due to the electrostatic interaction between ions and membranes (Petersen, 1993).

Most of membrane is consists negative charged at membrane surface. The counter-ion concentration will attract and present in the membrane phase at a higher level than that of co-ion. These results of electrostatic attractions and repulsions between the fix membrane charge and ionic species in the solution create a Donnan potential. The Donnan potential prevents the diffusion of the counter-ion from the membrane phase to the solution and the diffusion of co-ion from the solution to the membrane phase. When a pressure is applied to force the solution through the charged membrane, the effect of the Donnan potential is to repel the co-ion from the membrane. Because of an electroneutrality must be maintained in the solution, the counter-ion is also rejected (Williams, 2003). Donnan equilibrium models assume that a dynamic equilibrium is established when a charged membrane is placed in a salt solution (Bhattacharyya and Cheng, 1986; Bhattacharyya and Williams, 1992c).

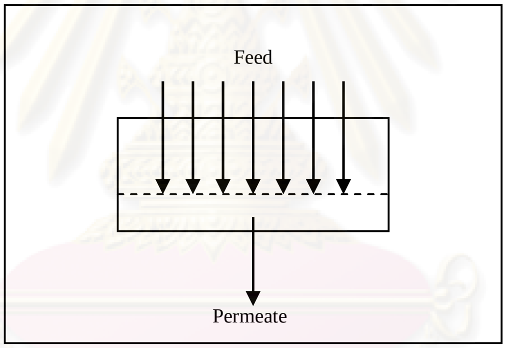
This model can be predicted that the rejection of solute was a function of effective charge density of membrane, solute concentration in external solution, and valence of solute. However, the two important factors for charge membrane including solute diffution and convective fluxes were not taken into account in this model.

2.2.3 System design

The system for membrane filtration can be categorized into 2 types: dead-end operation and cross-flow operation. All the feed is driven through the membrane, which implies that the concentration of rejected components in the feed increases and consequently the quality of the permeate decreases with time (Thanuttamavong, 2002).

2.2.3.1 Dead-end filtration

In dead-end filtration, all the water that enters the membrane surface is passed through the membrane. Some components will stay on membrane surface while water flows through. Figure 2.7 is showed the dead-end operation.

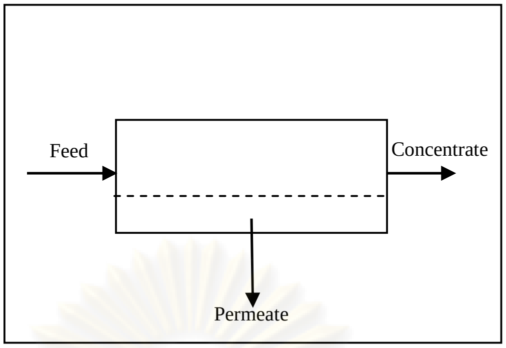


(Source: Adapted from Thanuttamavong, 2002)

Figure 2.7 Dead-end operation

2.2.3.2 Cross-flow filtration

In cross-flow filtration, the feed flows parallel to the membrane surface. Small part of the feed water is passing through the membrane; the largest part will leave the module. In many case, a cross-flow operation is preferred because of the rate of fouling is lower than in dead-end operation.



(Source: Adapted from Thanuttamavong, 2002)

Figure 2.8 Cross-flow operation

The cross-flow operation can be separated into two types: the single-pass method and the recirculation method. The schematic diagram of both methods is showed in Figure 2.9. In the single-pass method, the feed solution passes only once through the module, then the volume of feed solution decreases with time of operation, whereas, the recirculation method, the feed solution is sent back to pass module at several times. In this case, flow velocity and pressure can be adjusted to reduce the pressure drop and also minimize fouling.

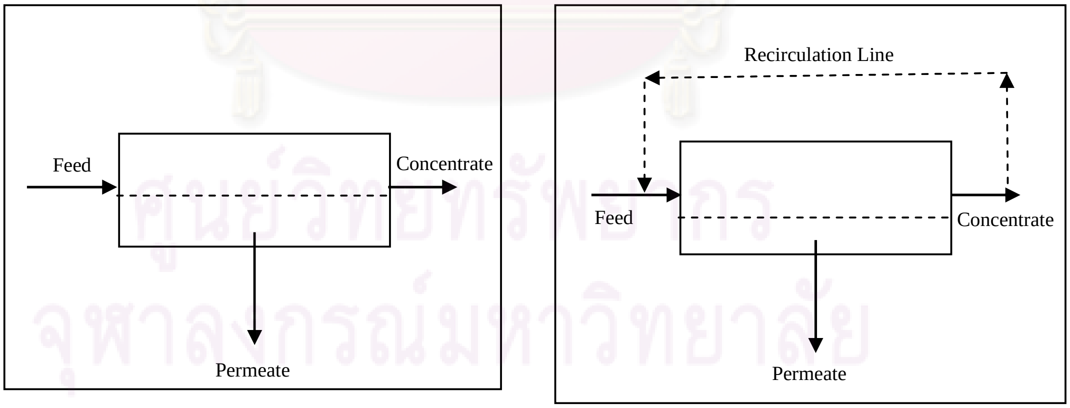
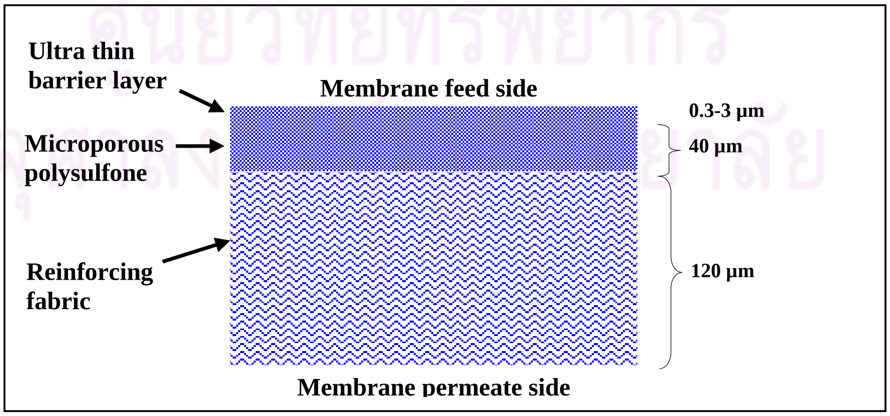


Figure 2.9 Method of the cross-flow operation

2.2.4 Ultra low pressure reverse osmosis membrane

The ultra low pressure reverse osmosis membrane (ULPRO membrane) is a new technology for membrane filtration which can be operates at very low pressure conditions (0.1-0.5 MPa) whereas a typical RO membrane requires very high operating pressure (1.5-15 MPa). Most of ULPRO membrane is multi-layer thin film composite of polymer. The active membrane surface layer usually consists of negatively charged sulphone or carboxyl group. ULPRO membrane have been identified as energy saving membranes with effective rejecting salts, trihalomethane formation potential (THMFP), heavy metal, color, and all micro organisms. ULPRO membrane can provide a specific flux of more than 60 L/m^2-h·MPa at pressure range about 0.2-0.9 Mpa. This specific flux is about 2 times of the specific flux of current generations of composite reverse osmosis membranes (Ozaki et al., 2000).

Petersen, 1993 reported that a thin film composite membrane may be defined as a bi-layer film formed by a two-step process. Such a membrane typically consists of a thick, porous, nonselective layer formed in the first process step, which is subsequently overcoated with an ultra thin barrier layer on its top surface in a second process step. The two layers are always different from one another in chemical composition. A typical thin film composite membrane as generally produced today is shown schematically in Figure 2.10.



(Source: Adapted from Robert, 1993)

Figure 2.10 Cross section of thin film composite membrane

A base layer of a woven or a non-woven fabric (for handling strength) is overcoated with a layer of an anisotropic micro porous polymer (usually polysulfone). The surface of the micro porous support is coated with an ultra thin polymeric composition, which provides the controlling properties as semi-permeability (Petersen, 1993).

Petersen, 1993 concluded that each individual layer can be optimized for its particular function, i.e. the ultra thin barrier layer can be optimized for the desired combination of solvent flux and solute rejection, while the porous support layer can be optimized for maximum strength and compression resistance combined with minimum resistance to permeate flow. Moreover, various chemical compositions can be formed into ultra thin barrier layers, including both linear and cross-linked polymers. The ability to generate an ultra thin layer in situ on a micro porous substrate also allows one to generate and use several of the cross-linked polymeric compositions, which can exhibit superior hydrophilic (viz. higher water permeability) and superior chemical resistance compared to linear polymeric compositions.

2.2.5 Specification of UTC-70 membrane

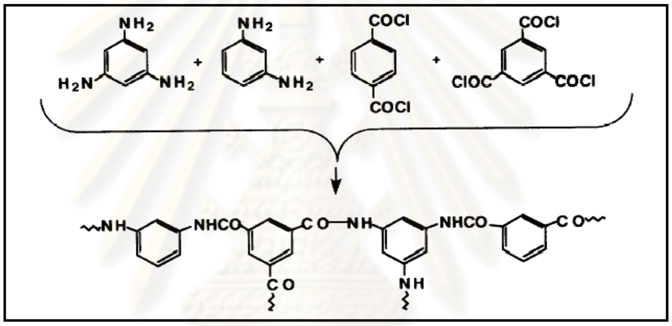
Some specifications of the UTC-70 membrane, which is ultra low pressure reverse osmosis membrane reported by Kurihara in 2003 are shown in Table 2.5.

Table 2.5 Specifications of UTC-70 membrane

	UTC-70 membrane
Material	Crosslinked Aromatic Polyamide
Structure	Thin film composite membrane
Rejection	Low MW Organic materials, Monovalent ions
MWCO	MW $\underset{\sim}{~}$ 60 [a]
Mechanism	Electric repulsion Solution diffusion Molecular interaction
Pore size	< 1 nm

(Source: Kurihara, Toray Company, 2003) [a] Yashinari, 1999

Petersen, 1993 reviewed that UTC-70 membrane was developed by Toray Corporation, Japan and was the basis of their SU-700 series of spiral element products. This membrane contains an aromatic polyamide barrier layer consisting of a blend of diamine and triamine interfacial reacted with a blend of diacyl and triacyl halides. The diamine is 1, 3-benzenediamine and the triamine appears to be 1, 3, 5-benzenetriamine. The triacyl halide is apparently trimesoyl chloride, and the diacyl halide, terephthaloyl chloride. The probable chemistry of UTC-70 membrane is given in Figure 2.11.



(Source: Toray Industries)

Figure 2.11 Probable chemistry of UTC-70 membrane

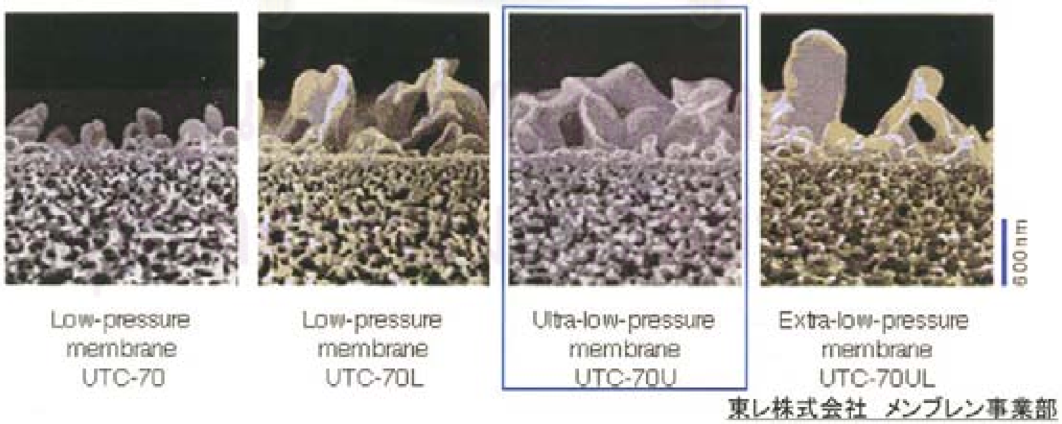


Figure 2.12 surface of UTC-70 membrane

2.2.6 Osmotic pressure

Osmosis is the movement of a solvent through a semi-permeable membrane from the more dilute solution side to the more concentrated solution side (C.N. Sawyer et al., 2003). The pressure that solutions need to reach equilibrium of both sides is call osmotic pressure (π).

Van't Hoff Equation can be use to determine of the osmotic pressure in a single salt solution, which can be calculated as follows:

$$\pi = i\,.z\,.nRT \tag{2-5}$$

where i = the permeation factor, z = the sum of the valency of the electrolyte, n = the salt concentration (mol/kg), R = the universal gas constants, and T = the temperature (^{o}K)

2.2.7 Concentration polarization

Concentration polarization is an undesirable phenomenon, which cause by the accumulation of retained solutes on surface of membrane. This phenomenon was a cause of precipitation or formation of a gel layer on membrane surface when concentration of solute exceeded its saturation limit (Matthiasson, 1980) and can elevate solute concentration on membrane surface and lead to decrease in rejection rate. Concentration polarization is an undesirable phenomenon as it exposes the membrane surface to an enhanced solute concentration. These acts to decrease permeate flux by reduction of the pressure driving force through the increase in the counteracting osmotic pressure (Sutzkover et al., 2000). Figure 2.13 shows the concentration profile of a solute in a concentration polarization model.

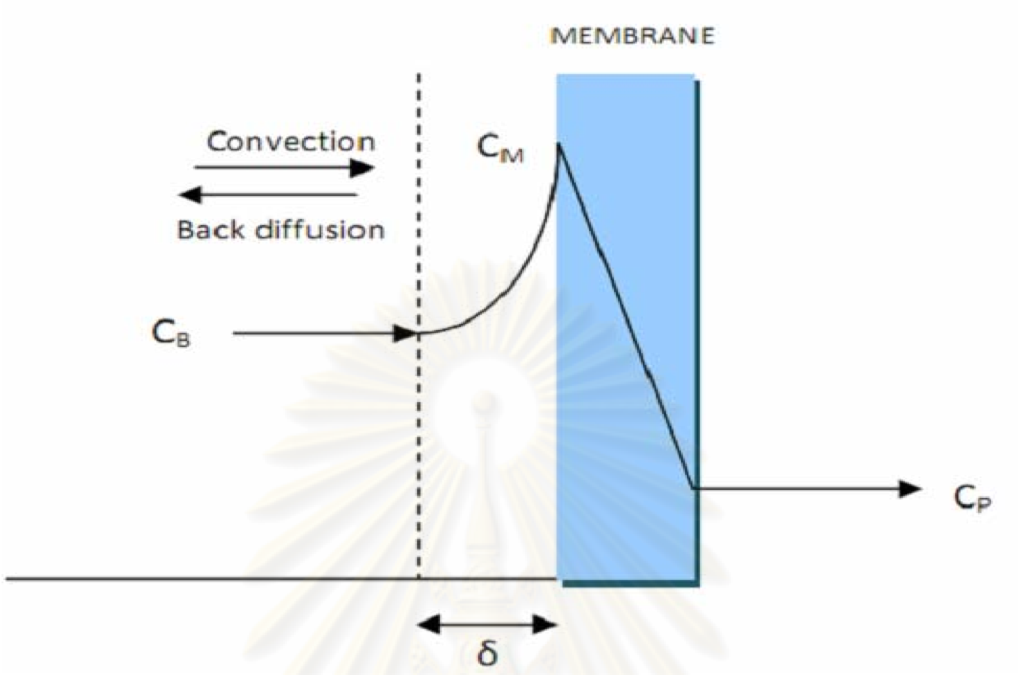


Figure 2.13 Concentration polarization model

Solutes are carried to the membrane by the bulk motion of fluid (convection). As water permeates through the membrane, the rejected solutes are left behind to accumulate at the membrane surface. The rejected solutes diffuse away from the membrane surface due to the resulting concentration gradient (bulk diffusion) (Bader, M.S.H., 2006).

Einar Matthiasson and Björn Sivik (1980) summerized negative aspects associated with concentration polarization were:

- An increase of chemical potential at the surface reduces the driving force for the filtration.
- An increase of hydrostatic resistance when the wall concentration of solute reaches the satuation limit. It is a cause of precipitation or formation of a gel on membrane surface.
- High solute concentration at membrane interface increases the risks for changes in composition of the membrane material due to chemical attack.

- Solute deposition on membrane surface can change the separation characteristic of the membrane.

According to the thin film model, the equations that use to describe the solute concentration on the membrane surface are given by;

$$\left(J_v\right)_{solute} = k\ln\left(\frac{C_M - C_P}{C_B - C_P}\right) \tag{2-6}$$

Where k = $\left(D/\delta\right)$= mass transfer coefficient, D = solute diffusion coefficient, δ = boundary layer thickness, $\left(J_v\right)_{solute}$ = permeate flux of solute solution, C_M = solute concentration prevailing on membrane surface, C_P = solute concentration in permeate solution, and C_B = solute concentration in bulk solution.

From the studied of Sutzkover, I. et al., 2000 in a simple technique for determining the mass transfer coefficient and the concentration polarization level in a reverse osmosis (RO) system. The mass transfer coefficient was estimated by following equation.

$$k = \frac{\left(J_v\right)_{solute}}{\ln\left\{\frac{\Delta P}{\pi_B - \pi_P}\cdot\left[1 - \frac{\left(J_v\right)_{solute}}{\left(J_v\right)_{H_2O}}\right]\right\}} \tag{2-7}$$

Where $\left(J_v\right)_{H_2\mathrm{O}}$ = permeate flux of pure water, ΔP = transmembrane pressure, π_B = osmotic pressure of bulk solution, and π_P = osmotic pressure of permeate solution.

It was assumed that solute flow through membrane was a diffusion controlled. From the solution-diffusion flow model, the permeate flux of a solute solution $(J_v)_{solute}$, and solute flux, J_i, through membrane is given by

$$J_v = k_w \left(\Delta P - \Delta \pi \right) \tag{2-8}$$

where k_w = pure water permeability.
ΔP = transmembrane pressure
$\Delta \pi$ = osmotic pressure

$$J_i = k_i \left(C_M - C_P \right) \tag{2-9}$$

Where k_i = solute mass transfer coefficient.
C_M = concentration of membrane surface
C_P = concentration of permeate

2.2.8 Membrane fouling

2.2.8.1 Background

Membrane fouling is a problem in membrane separation processes. Membrane fouling is referred to a flux decline of membrane filter that caused by accumulation of certain components in feed water on membrane surface or in membrane matrix (Liu et al., 1998). Membrane fouling is also cause of the quality of permeate declined and membrane degradation. According to the type of fouling, membrane fouling can be categorized into 4 types: inorganic fouling/scaling, particles/colloids fouling, microbial/biological fouling, and organic fouling (Liu et al., 1998).

- Inorganic fouling/ scaling

Inorganic fouling and scaling are caused by the accumulation of the inorganic precipitates (i.e. metal hydroxides and scales) on the membrane surface or within membrane matrix (Liu, C. et al., 1998). The inorganic soluble compounds will precipitate on the membrane surface and form a scale layer when the concentrations of these compounds exceed the solubility limit (R. P., Schneider et al., 2005).

- Particles/ colloids fouling

Particles and colloids represent a major type of foulants present in most feed waters (S.G. Yiantsios and A.J. Karabelas, 1998). During filtration processes, particles and colloids retained by a membrane will form a cake layer on the membrane surface. The fouling rates and resistance of this cake layer can be significantly affected by the colloidal interactions, which are described in the colloid science (Lianfa Song and Gurdev Singh, 2005).

- Microbial/ Biological fouling

Microbial fouling is the attachment and growth on the membrane surface of microbes, which leads to the formation of biofilms (W.G. Characklis and K.C. Marshall, 1990). When these microbes attach to the membrane, they will produce extracellular polymetric substances (EPS) to form a viscous, slimy, and hydrated gel on that membrane (Liu, C. et al., 1998). Biofouling can significantly reduce permeate flux and permeate quality (L.Y. Dudley and J.S. Baker).

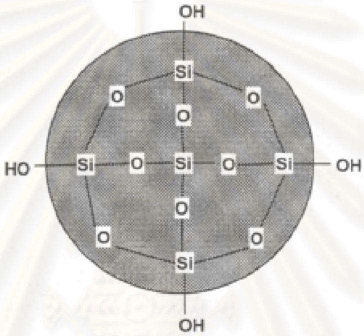
- Organic fouling

Organic fouling is dominant in membrane filtration process with feed water containing relatively high natural organic matters (NOM). Generally, surface water such as river and lake contain higher NOM than groundwater. For feed water containing high NOM, it is the most significant factor contributed to flux decline (Mallevialle et al., 1989; Lahoussine-Turcaud et al., 1990)

2.2.8.2 Silica fouling

Silica $(SiO_2)_n$, is widely found in the environment. In natural water sources, silica is released from the weathering of minerals, which containing silica species such as asbestos, feldspar, mica, clay and even opal (Darton, 1999). Many factors are influenced the formation of silica including temperature, pH and metal ions.

The silica $(SiO_2)_n$ structure is shown in Figure 2.13. Thus, the molecules of silica represented by the formula SiO_2 is polymeric form, and is more accurately represented by the formula of $(SiO_2)_n$ where n is infinited in number, allowing for extensions in term of amorphous and crystalline forms of silica (R. Y. Ning et al., 2005).



(Source: K.D. Demadis et al., 2005)

Figure 2.14 Silica $(SiO_2)_n$ structure

Silica forms are categorized into three types depending on their sizes: dissolved silica or reactive silica (< 0.001 μm), colloidal silica (0.001 – 1 μm), and particulate silica (> 1 μm), respectively. The silica solubility of 120 mg/L is valid for pH values from 6 to 8 and increases as pH value increases (Rodriguez, 2005).

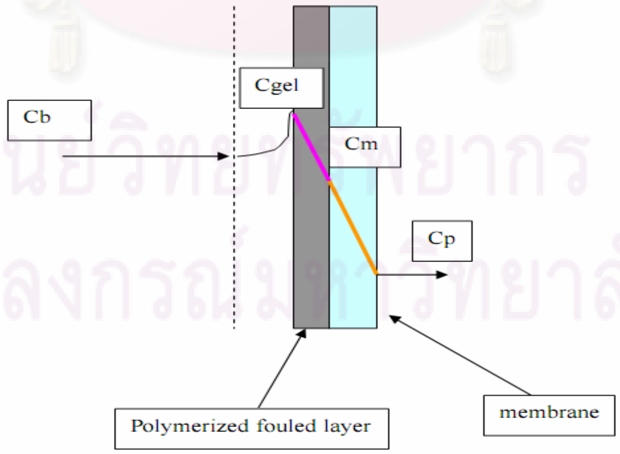
Silica is one of the foulants present in the feed water in RO membrane plants. Silica fouling has been described as a major unsolved problem in desalination units (Sheikholeslami, R., and Tan, S., 1999). Silica fouling can reduce permeate flux in membrane filtration processes. The silica in groundwater in Chiang Mai Basin has relatively low with an average concentration of 28.0 mg/L, a long-term operation of RO/NF membranes may accumulate silica on membranes if no measure is taken to control silica (Matsui, 2007). The pretreatment process can not removed silica concentration, then it can be effect in reverse osmosis process. The solubility of SiO_2 is 100~120mg/L at pH7 and 25℃ (Matsui, 2006).

Matsui, 2007 studied the effect of CaF, $CaCo_3$ and SiO_2 on groundwater defluoridation. The result indicated that SiO_2 in feed water caused the maximum flux decline among 3 factors. Silica deposits are difficult and costly to remove and not easily controlled by anti-scalants (Hasson, 2005).

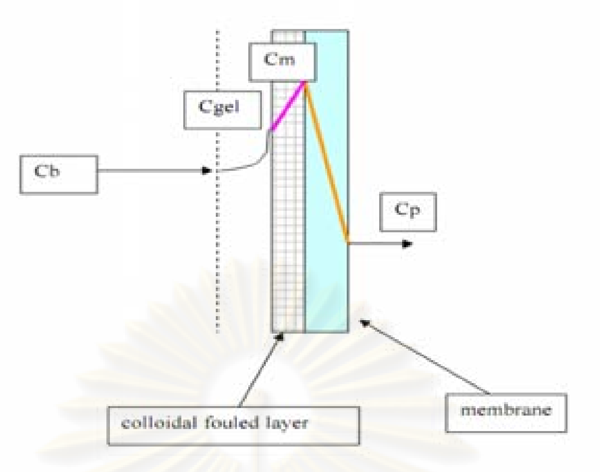
Sahachaiyunta reported that the type and extent of silica fouling depends on the condition of the system such as the silica concentration, pH of feed solution, temperature, and presence of other species.

There are two relevant categories of fouling, namely precipitation fouling and particulate fouling. Precipitation fouling occurs when monomeric silica polymerizes at the membrane surface, also called scaling or polymerized silica fouling. On the other hand, particulate fouling involves the accumulation of colloids that are formed initially in bulk solution and deposited subsequently on the membrane surface (Sahachaiyanta, P. et al., 2002).

From the concentration polarization model, figures 2.15 show the schematic profile of membrane fouling.



(a) Polymerized fouled layer



(b) Colloidal fouled layer

Figure 2.15 The schematic model of polymerized and colloidal fouled layer

CHAPTER III

METHODOLOGY

3.1 The study area

The study area is in Chiang Mai basin, Chiang Mai Thailand. The Chiang Mai basin covers about 3,000 km^2 of Chiang Mai and Lamphun province. This area situated between 17-19 latitude N and 98-99 longitude E. It has 35 km wide in NNE-SSW direction and 140 km length in N-S direction (Matsui, 2007). The ping river runs through the basin between Lamphun province and Chiang Mai province. The area is divided into the flood plain, the low terrace, the high terrace, and the mountain area. Fluoride in groundwater was enriched in aquifers of alluvial plains in the northern part of Lamphun Province (Matsui, 2006). The possible causes of high fluoride in groundwater are the presence of fluoride mineral and mixing of hot spring water. Groundwater from two sites in Chiang Mai basin was selected as sampling point. The sampling point is shown in Figure 3.1

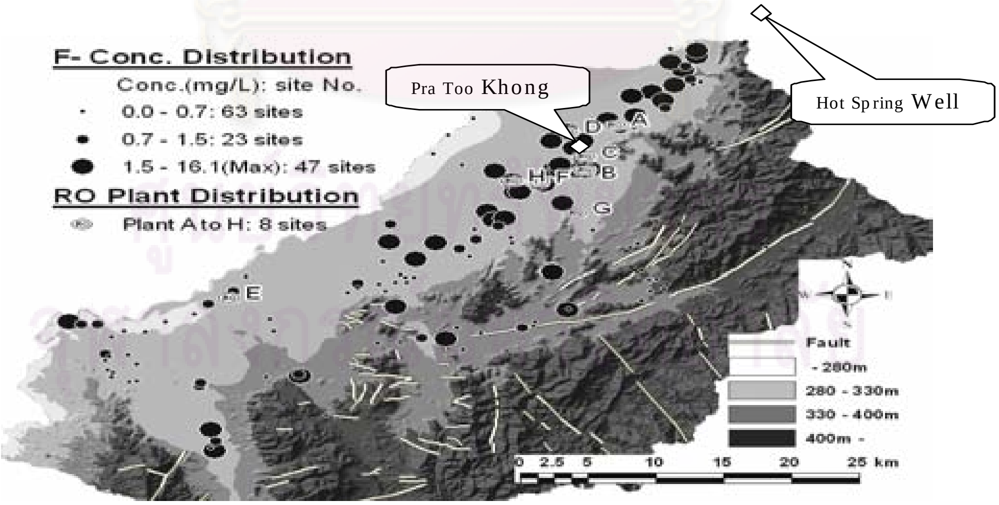


Figure 3.1. Chiang Mai basin and sampling points

The first site was Pra Too Khong, Tambon Ban Klang, Muang district, Lamphun province, which is defined as a very high fluoride concentrations site.

Figure 3.2 describe the position of Pra Too Khong Bottled Drinking Water Plant. This site was established by a private company to produce the bottled drinking water for people in Tambon Ban Klang. This plant has a capacity of 5 m^3/day. Groundwater in Tambon Ban Klang is strongly recommended that could not be used directly as drinking water. The groundwater which passed through the pre-treatment was collected as water samples for membrane experiment. The pre-treatment can remove ferrous ions which can be oxidized and clog the membrane. This phenomenon reduced a permeate water flux and finally, the membrane could not work properly anymore (Aunnop, 2006).

Figure 3.2 Pra Too Khong, Tambon Ban Klang, Muang district, Lamphun province

The pre-treatment of Pra Too Khong drinking water plant consists of three processes. First is de-ironed facility to remove iron and manganese. The next is activated carbon to remove color odor and organic substance. The last is cation exchange resin to remove calcium, sodium, magnesium and potassium. Groundwaters, which pass the pre-treatment was collected and used as feed water in the experiment. From this point, Pra Too Khong groundwater mean pretreated water from Pra Too Khong drinking water plant. The groundwater characteristics of raw water and pretreated water from Pra Too Khong drinking water plant are shown in Chapter 4.

The second site was San Kam Pang hot spring well, Tambon Ban Sahakorn, Mae-on district, Chiang Mai province. This site is defined as high fluoride concentration. The hot water might mix with nearby groundwaters, thus a mixed groundwater would contain high fluoride concentration (Aunnop, 2007). Kundu *et al.* (2001) reported that fluoride in groundwater has derived from the intrusion of fluoride rich geothermal water. This site also contains high silica concentrations. Aunnop (2007) reported that groundwater from hot spring contains silica concentration 147.1 mg/L. The groundwater characteristics of San Kam Pang hot spring are shown in Chapter 4.

Figure 3.3 San Kam Pang hot spring well, Chiang mai province

3.2 Materials

3.2.1 Feed Tank

A plastic container with 10 L capacity was used to contain the feed solution.

Figure 3.4 Feed tank

3.2.2 Temperature-controlling water bath

In this study, temperature was controlled at 25 ± 5 °C by using a temperature-controlling water bath, which could control temperature in range of 0 to 70°C

Figure 3.5 The temperature-controlling water bath

3.2.3 Membrane module

The membrane module is the C10-T module, which cross-flow membrane unit, was obtained from the Nitto Denko Corporation, Japan. The membrane module and equipment in membrane module are shown in Figure 3.6 and 3.7.

Figure 3.6 membrane module

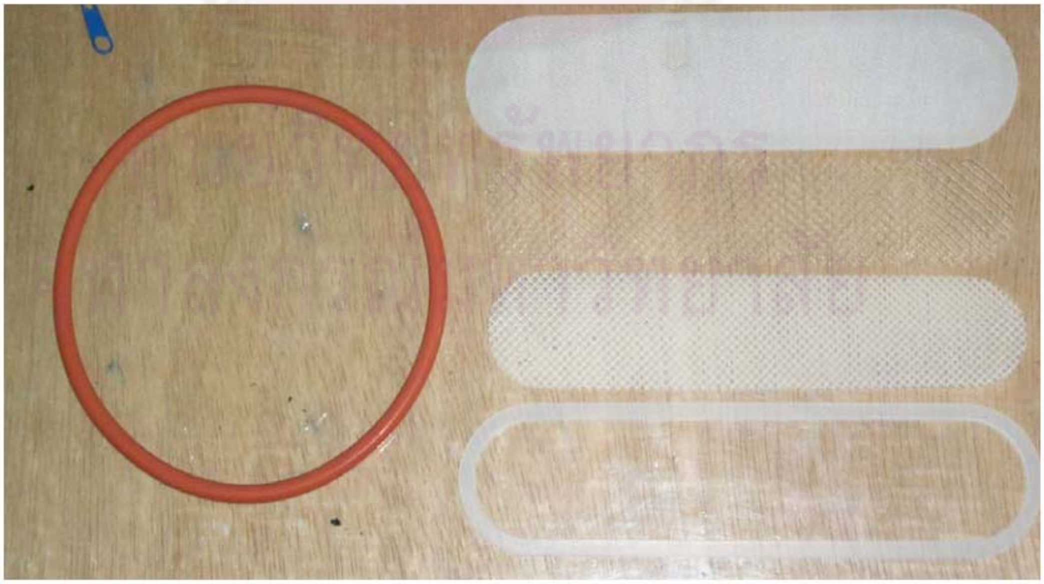


Figure 3.7 Equipment in membrane module

3.2.4 Membrane

The flat sheet of UTC-70, which ultra low pressure reverse osmosis membrane (ULPRO membrane), was obtained from the Toray Corporation, Japan. It can provide 60 cm^2 of filtration area. The UTC-70 flat sheet is shown in Figure 3.8.

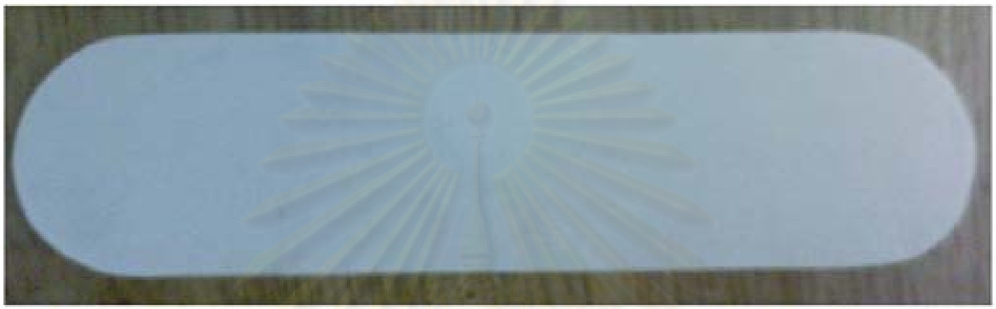


Figure 3.8 Flat sheet of UTC-70 membrane

3.2.5 Valve

The concentrated valve was set in membrane experiment system, which used to adjust the pressure in the system.

3.2.6 Flow meter

The flow meter was set in membrane experiment system and used to detect the water flow through the membrane experiment system.

3.2.7 Pump

The gear pump was obtained from the Iwaki Company, Japan. It could be operated at maximum operating pressure of 0.55 MPa, and a maximum capacity of 2.0-2.4 liters per minute.

3.2.8 Fluoride

Fluoride concentration in concentration polarization experiment was prepared by using sodium fluoride (J.T. Baker, USA.).

3.2.9 Silica

Silica concentration was prepared by using sodium metasilicate nonahydrate (Wako Pure Chemistries, Ltd., Japan).

3.3 Membrane experiment

3.3.1 Water sampling

About 20 L of groundwater from both sites was collected in plastic tank and carried to the laboratory of the Department of Environmental Engineering, Faculty of Engineering, Chiang Mai University.

3.3.2 Membrane experimental procedure

This research was conducted in a cross-flow operation unit. The membrane module (C10-T Module, Nitto Denko Corp.) with a flat sheet of UTC-70 ULPRO membrane was used in the experiment. The transmembrane pressure was varied at 0.3 and 0.5 MPa. The temperature was controlled at 25°C by using a temperature-controlling water bath. The diagram of the membrane experiment process is shown in Figure 3.9 and Figure 3.10.

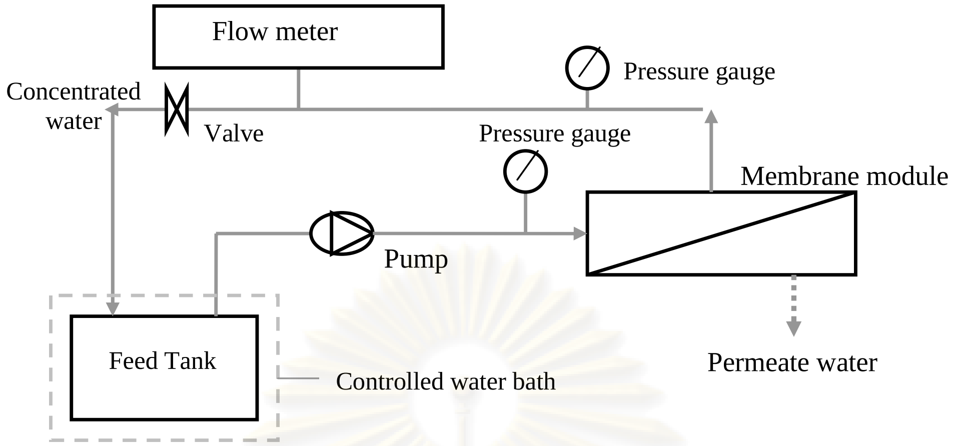


Figure 3.9 Diagram of membrane experiment

Figure. 3.10 The membrane experiment

3.3.3 Preparation of the membrane experiment

The ULPRO membrane (UTC-70 membrane) was prepared by using the following procedure.

1. The equipment in module was clean with milli-Q water.

2. The flat sheet UTC-70 membrane was cut and washed in milli-Q water. Then it was set up properly in the cell unit.

3. It was put through the module for 1 hour under pressure 0.30 MPa with 1 L of milli-Q water for washing the membrane.

4. Concentrated water and permeate water were recycled to the feed tank.

Figure 3.6 show the preparation of the membrane experiment with 1 L of milli-Q water.

Figure 3.11 The preparation of membrane experiment

3.3.4 Concentration polarization experiment

In the concentration polarization experiment. The feed solutions were prepared by using the milli-Q water combined with varied NaF concentration (0, 10, 25, and 50 mM).

1. 5 L of feed solution in the feed tank was put through the membrane experiment process. The electroconductivity and pH of feed solution was recorded. The concentrated water was recycled to the feed tank.

2. The permeate flux was observed every 10 minutes in the first hour and every 30 minutes in the next hour. The permeate flux was determined by using 10 ml measuring cylinder to identify the time at the permeate water reached 10 ml.

3. At 250 ml of permeate water obtained, the concentrated water and permeate water were collected in 120 ml plastic bottle for analyzed the fluoride concentration. The electroconductivity and pH were measured.

4. The membrane experiments for other NaF concentration were done by starting with the preparation of the membrane experiment step.

5. At the end of the membrane experiment, the employed membrane was replaced by the new membrane sheet.

6. The membrane experiments for other transmembrane pressure were done.

The concentration polarization experiment condition is shown in Table. 3.1

Table 3.1. Concentration polarization experiment condition

Runs no.	Transmembrane pressure (Mpa)	NaF concentration (mM)
A1	0.3	0
A2		10
A3		25
A4		50
B1	0.5	0
B2		10
B3		25
B4		50

3.3.5 Groundwater defluoridation experiment

In the groundwater defluoridation experiment, groundwater from two sites was used as feed solution.

1. 5 L of groundwater from site 1 (Pra Too Khong) was filtrated with 0.45 μm filter and put through the membrane experiment process. The electroconductivity and pH was recorded. The concentrated water was recycled to feed tank.

2. The permeate flux was observed every 10 minutes in the first hour and every 30 minutes in the next hour. The permeate flux was determined with the same method in concentration polarization experiment.

3. At 250, 500, 750, and 1000 ml of permeate water obtained, the concentrated water and permeate water were collected in 120 ml plastic bottles. The electroconductivity and pH were measured.

4. At the end of the membrane experiment, the employed membrane was replaced by the new membrane sheet.

5. The membrane experiment for other transmembrane pressure was done.

6. The membrane experiments for groundwater from site 2 (San Kam Pang hot spring well) were done by starting with the preparation of the membrane experiment step and continued with step 1.

3.3.6 Silica fouling experiment

In this experiment, silica concentration 500 mg/L was added to groundwater to prepared feed solutions.

1. 5 L of feed solution was put through the membrane experiment process. The electroconductivity and pH of feed solution was recorded. The concentrated water was recycled to feed tank.

2. The permeate flux was observed every 10 minutes in the first hour and every 30 minutes in the next hour. The permeate flux was determined with the same method in concentration polarization experiment.

3. At 250, 500, 750, and 1000 ml of permeate water obtained, the concentrated water and permeate water were collected in 120 ml plastic bottles. The electroconductivity and pH were measured.

4. At the end of the membrane experiment, the employed membrane was replaced by the new membrane sheet.

5. The membrane experiment for other transmembrane pressure was done.

6. The experiments for other feed solution were done by starting with the preparation of the membrane experiment step and continued with step 1.

Table 3.2 demonstrated the groundwater defluoridation and silica fouling experiment condition.

Runs no.	Transmembrane pressure (Mpa)	Groundwater site	Silica adeed (mg/L)
1	0.3	Pra Too Khong	0
2	0.3		500
3	0.5		0
4	0.5		500
5	0.3	San Kam Pang hot Spring	0
6	0.3		500
7	0.5		0
8	0.5		500

3.4 Analytical method and instruments

3.4.1 Fluoride

The fluoride concentration in feed solution, concentrated solution and permeate solution was analyzed in accordance with standard method 4110; section 4110B by Ion-Chromatography with Chemical Suppression of Eluent Conductivity.

3.4.2 pH

The pH of concentrated water and permeate water was measured by a Horiba pH meter, Model D-13E with an accuracy of ± 0.01 pH unit.

3.4.3 Electrical Conductivity (EC)

The electrical conductivity (EC) of concentrated water and permeate water was measured by a WTW electrical conductivity meter, Model Cond 330i.

3.4.4 Temperature

Temperature of feed water, concentrated water and permeate water were measured by thermometer.

CHAPTER IV

RESULTS AND DISCUSSION

The results of membrane experiments and their analysis for each part of experiments were presented in the following pages.

4.1 Concentration polarization experiment

The concentration polarization was studied. The feed solution was varied the fluoride concentration at 0, 10, 25, and 50 mM by dissolving NaF in milli-Q water.

4.1.1 Permeate water flux

In this experiment, the permeate water flux was calculated as followed:

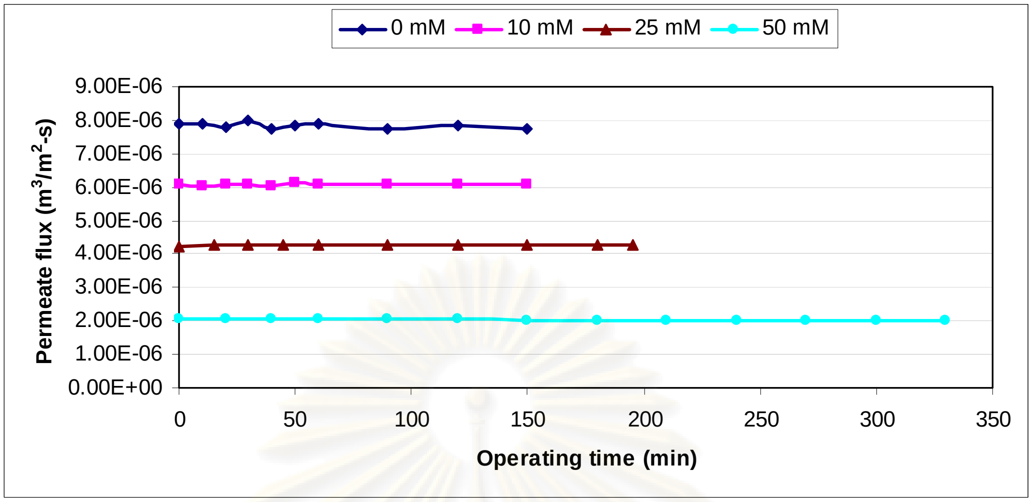
$$\text{Permeate water flux } (m^3/m^2 \cdot sec) = \frac{\text{10 mL of permeate water}}{A \times T} \tag{4-1}$$

Where A = surface area of UTC-70 membrane (60×10^{-4} m^2)

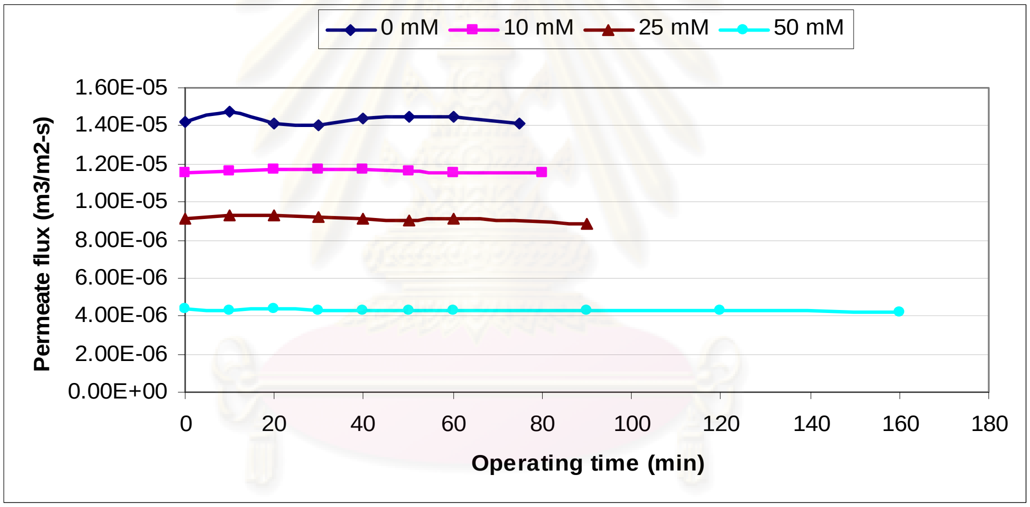
T = Time when the permeate volume reached 10 ml

The operating time that used to reached 250 ml of the permeate water volume were different. It depends on the operating transmembrane pressure. The operating time of varied fluoride concentration (0, 10, 25, and 50 mM) in feed solutions was 150-540 minutes and 80-270 minutes at 0.3 and 0.5 MPa, respectively. The results demonstrated that the highest operating transmembrane pressure (0.5 MPa) provide the shortest operating time.

The permeate water flux as a function of operating time until 250 ml of permeate water obtained are shown in Figure 4.1.



(a) Permeate flux at 0.3 MPa



(b) Permeate flux at 0.5 MPa

Figure 4.1. The permeate flux as a function of operating time.

The permeate flux at 250 ml permeate water obtained under operating transmembrane pressure 0.3 and 0.5 MPa was investigated. The permeate flux under 0.3 MPa were 7.899x10^{-6}, 6.061x10^{-6}, 4.253x10^{-6} and 2.012x10^{-6} m^3/m^2-s at 0,10,25 and 50 mM, and those under 0.5 MPa were 1.41x10^{-5}, 1.157x10^{-5}, 8.818x10^{-6}, and 4.188x10^{-6} m^3/m^2-s at 0,10,25 and 50 mM, respectively.

Table 4.1 showed the relationship between permeate flux and operating transmembrane pressure at various fluoride concentrations.

Table 4.1 Permeate flux at 250 ml of permeate water obtained under 0.3 and 0.5 MPa

Feed concentration	Permeate flux (m^3/m^2-s)	
(mM)	0.3 MPa	0.5 MPa
0	7.899×10^{-6}	1.410×10^{-5}
10	6.061×10^{-6}	1.157×10^{-5}
25	4.253×10^{-6}	8.818×10^{-6}
50	2.012×10^{-6}	4.188×10^{-6}

From the results as shown in Table 4.1, it was found that the higher permeate flux were obtained at higher operating transmembrane pressure. It was indicated that when the operating transmembrane pressure increased, the permeate flux also increased. This result related to the studied of Bhattacharyya et al., 1992 and Williams, M.E., 2003, which reported that the operating pressure affected the performance of the membrane in the membrane separation processes.

The fluoride concentration in feed water also affected on the permeate flux. From the result, the permeate flux decrease with increasing of fluoride concentration in feed water due to the effect of osmotic pressure. The osmotic pressure was calculated from Equation (2-5). The results of osmotic pressure of each fluoride concentration are shown in Table 4.1. It can be stated that higher osmotic pressure could be obtained from lower fluoride concentration. Increasing bulk salt concentration causes a higher osmotic pressure drop, which results in a smaller permeate flux (Hoek, 2005). The performance of ULPRO membrane was evaluated by using sodium chloride. The results show that the flux is slightly linear with a negative slope in higher feed concentration (Ozaki et al, 2002).

The solute – diffusion model was used to describe the relationship between permeate flux and operating transmembrane pressure and osmotic pressure by following equation.

$$J_v = k_w\left(\Delta P - \Delta\pi\right) \tag{4-2}$$

where k_w = pure water permeability

ΔP = transmembrane pressure

$\Delta\pi$ = osmotic pressure

From Equation (4-2), the operating transmembrane pressure (ΔP) and osmotic pressure ($\Delta\pi$) are involved the value of permeate flux. The permeate flux increase with the increasing operating transmembrane pressure and decreasing of the osmotic pressure.

4.1.2 The mass transfer coefficient (k)

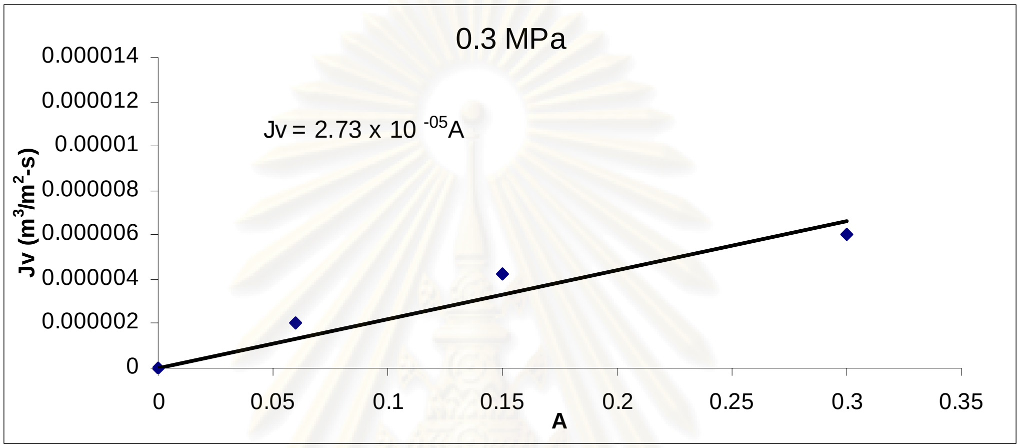
The mass transfer coefficient in solution of each operating transmembrane was investigated.

The fluoride concentration in feed water, concentrated water and permeate water was analysed by ion-chromatography. While, the osmotic pressure for each parts were calculated from Equation (2-5). The results are shown in Table 4.1.

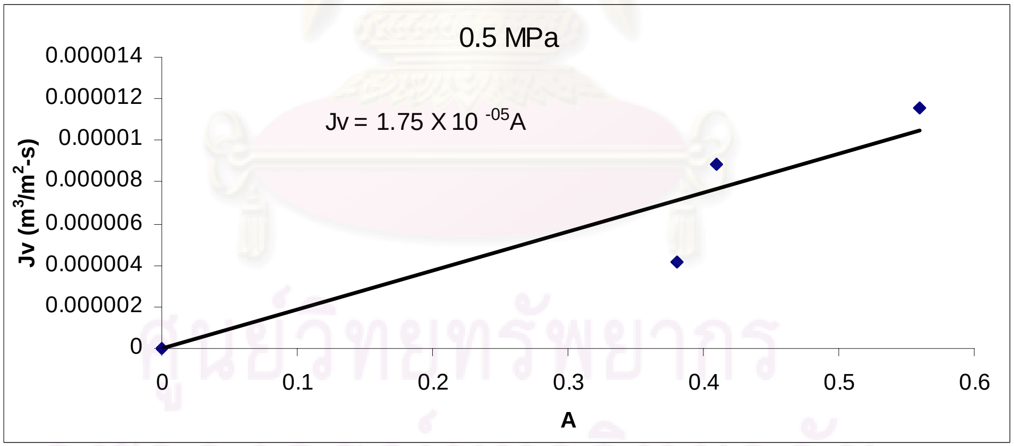
From the following equation, the mass transfer coefficient was estimated.

$$k = \frac{(J_v)_{solute}}{\ln\left\{\dfrac{\Delta P}{\pi_B - \pi_P}\cdot\left[1-\dfrac{(J_v)_{solute}}{(J_v)_{H_2O}}\right]\right\}} \tag{4-3}$$

The mass transfer coefficient for each transmembrane was obtained by plotting between $(Jv)_{solute}$ and A. (A= $\ln\left\{\frac{\Delta P}{\pi_B-\pi_P}\cdot\left[1-\frac{(J_v)_{solute}}{(J_v)_{H_2O}}\right]\right\}$) and the resuls are shown in figure 4.2



(a) Average mass transfer coefficient at 0.3 MPa



(b) Average mass transfer coefficient at 0.5 MPa

Figure 4.2 Average mass transfer coefficients

From Figure 4.2, it was found that the mass transfer coefficient was $2.73\text{x}10^{-5}$ and $1.75\text{x}10^{-5}$ m/s at 0.3 and 0.5 MPa, respectively. As a result, it was indicated that the operating transmembrane pressure was affected on the mass transfer coefficient.

Mass transfer coefficient in water (*k*) depends on feed water pH, temperature, reynolds number. This *k* value can be utilized in some range of fluoride concentration, until the plot is straight line.

The mass transfer coefficient for 0.3 and 0.5 MPa was used in groundwater defluoridation experimental.

Table 4.2 The results of mass transfer coefficient and permeate flux

Run no.	Feed solution	Transmembrane pressure	Concentrated water (C_B)		Permeate water (C_P)		$\Delta\pi$	Permeate flux (m^3/m^2-s)
		(Mpa)	mM	π_B	mM	π_P	$\pi_B-\pi_P$	Jv x 10^{-5}
A1	0	0.3	0	0	0	0	0	0.789
A2	9.80		11.81	0.0545	0.60	0.0028	0.0518	0.606
A3	24.21		28.58	0.1320	2.83	0.0131	0.1189	0.425
A4	49.81		55.24	0.2551	9.74	0.0450	0.2101	0.201
B1	0	0.5	0	0	0	0	0	1.412
B2	9.94		11.39	0.0526	0.57	0.0026	0.0500	1.157
B3	24.57		28.00	0.1293	2.58	0.0119	0.1174	0.882
B4	51.47		55.75	0.2575	7.54	0.0348	0.2227	0.419

4.1.3 Concentration polarization.

The fluoride concentration on membrane surface (C_M) was calculated from Equation (2-6). The average mass transfer coefficient at 0.3 and 0.5 MPa was used. When the fluoride concentration on membrane surface was obtained, the concentration polarization level ($f = C_M/C_B$) was estimated. Moreover, the intrinsic rejection rate ($Rint = 1-C_P/C_M$) and observed rejection rate ($Robs = 1- C_P/C_B$) were calculated. The results are shown in Table 4.3.

Table 4.3 Level of concentration polarization

Run no.	Concentrated water (C_B) mM	Permeate water (C_P) mM	Membrane surface (C_M) mM	f	R_{int} %	R_{obs} %
A2	11.81	0.60	14.60	1.24	95.9	94.9
A3	28.58	2.83	32.92	1.15	91.4	90.1
A4	55.25	9.74	58.72	1.06	83.4	82.4
B2	11.39	0.57	21.52	1.89	97.4	95.0
B3	28.00	2.58	44.66	1.60	94.2	90.8
B4	55.75	7.54	68.78	1.23	89.0	86.5

From the results in Table 4.3, it was found that the fluoride concentration on membrane surface (C_M) was higher than in concentrated water (C_B) in all runs. Therefore, the intrinsic rejection rate was higher than the observed rejection rate too.

Moreover, the fluoride concentration in permeate water was increased when increasing the feed concentration. It could be described that at low feed concentration, electrostatic repulsion between solute and membrane was high. However, at high feed concentration, electrostatic repulsion between solute and membrane was decreased. As a result, it leaded to increase in solute concentration of permeate (Wongrueng et al., 2007).

From the results of the concentration polarization level for each feed fluoride concentrations, it was found that at lowest feed fluoride concentration (10 mM) in Run A2 and B2, the highest level of concentration polarization 1.24 and 1.89 was obtained at 0.3 and 0.5 MPa, respectively. The concentration polarization level was decreased with increasing of feed fluoride concentration. Wongrueng et al., 2007 reported that high concentration polarization level (*f*) was observed at low feed concentration. Furthermore, if the concentration polarization level was plotting with permeate flux at 250 ml permeate water obtained. It was found that the concentration polarization level as a function of permeate flux. The results are shown in Figure 4.3.

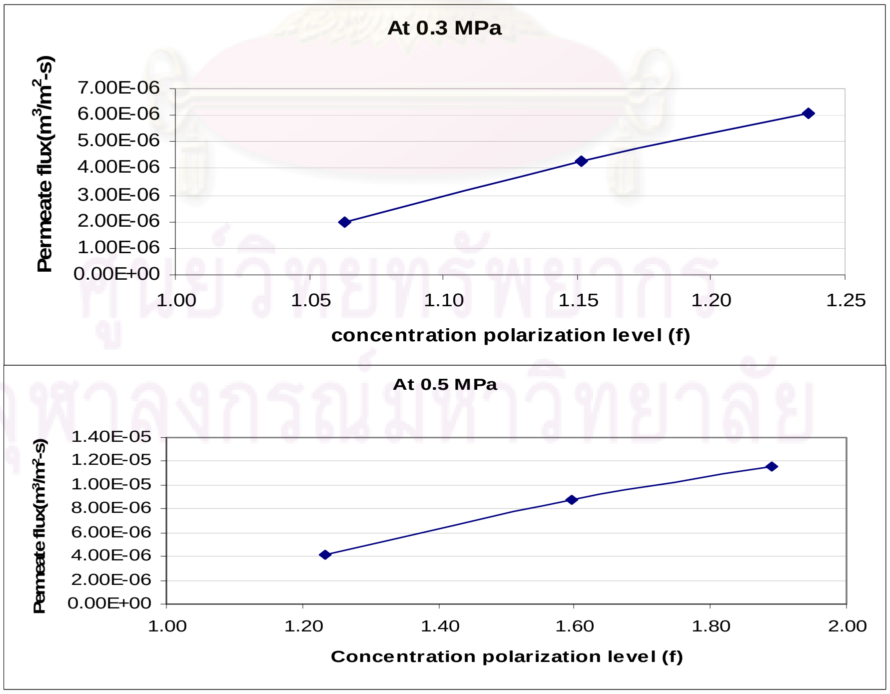


Figure 4.3 The relationship between concentration polarization level and permeate flux at 0.3 and 0.5 MPa

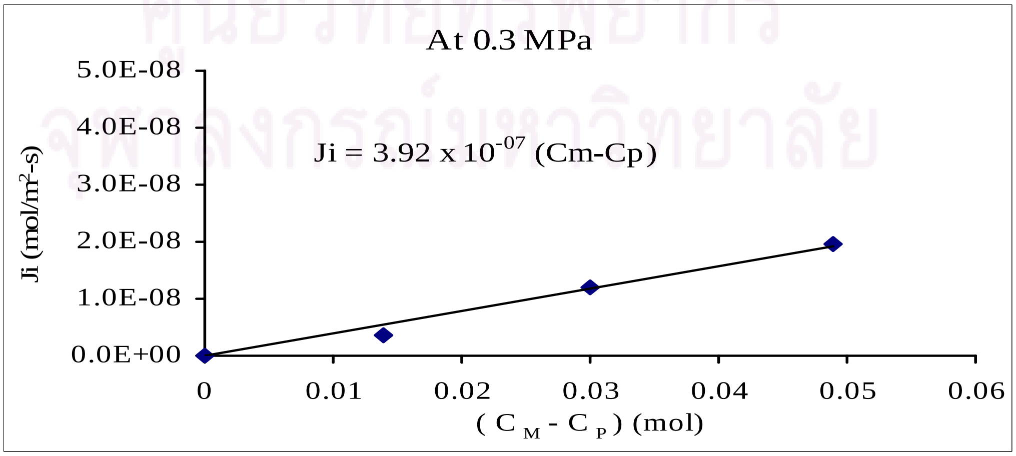
From Figure 4.3, it was found that the highest permeate flux was provided the highest concentration polarization level both of operating transmembrane pressure (0.3 and 0.5 MPa). At low feed concentration, high permeate flux could transport solute onto membrane surface more than low permeate flux, the higher concentration polarization level was obtained. Thus, besides feed concentration, permeate flux should be considered in a control of concentration polarization.

4.1.4 Solute mass transfer coefficient.

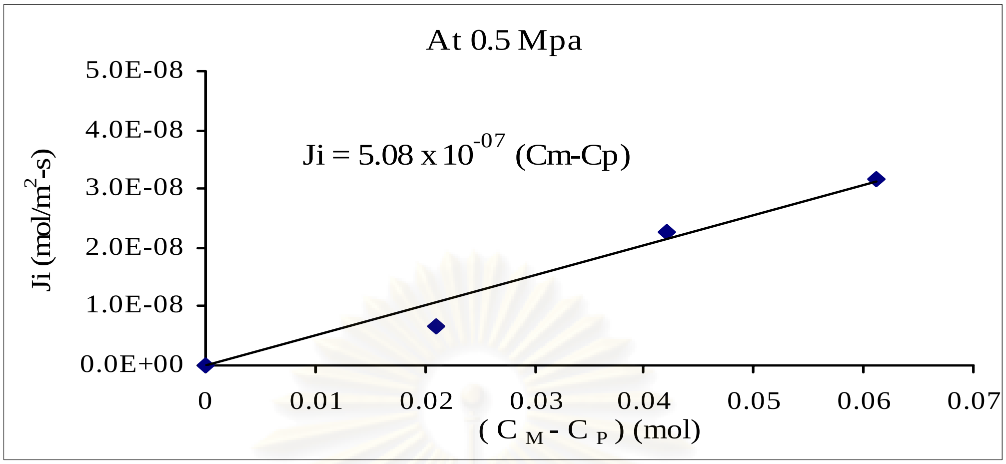
The solute mass transfer coefficient through the membrane was investigated. The solute flux through the membrane (*Ji*) was calculated from permeate flux and fluoride concentration in permeate water. Then the solute mass transfer coefficient (*Ki*) through UTC-70 membrane was estimated from Equation (4-4).

$$J_i = Ki(C_M - C_P) \tag{4-4}$$

From this equation, the solute mass transfer coefficient through membrane can estimate by plotting between solute fluxes (*Ji*) and (C_M-C_P). The results are shown in Figure 4.4.



(a) At 0.3 MPa



(b) At 0.5 MPa

Figure 4.4 Solute mass transfer coefficients

From Figure 4.4 the solute mass transfer coefficient through membrane was 3.92×10^{-7} and 5.08×10^{-7} m/s at 0.3 and 0.5 MPa, respectively. The average of solute mass transfer coefficient through membrane was 4.5×10^{-7} m/s. The average of solute mass transfer coefficient was used in groundwater defluoridation experimental.

4.2 Groundwater defluoridation

Groundwater from Pra Too Khong drinking water plant, Lamphun and San Kam Pang hot spring well, Chiang mai was collected and filtrated by UTC-70 ultra low pressure reverse osmosis membrane. The results are shown separately in each part.

4.2.1 Fluoride concentration in groundwater and permeate water flux

The fluoride concentration in groundwater from both sites was analyzed by ion-chromatography. The permeate flux was calculated with the same equation in concentration polarization experiment.

The fluoride concentration and permeate flux of each sites were shown separately in following parts

4.2.1.1 Pra Too Khong groundwater

In September, 2008, the performance of three membrane plants in Lamphun province including Pra Too Khong drinking water plant was investigated. At Pra Too Khong drinking water plant, raw water, pretreated water and permeate water were collected. The characterization of Pra Too Khong groundwater was analyzed and the results are shown in Table 4.4.

Table 4.4 Pra Too Khong groundwater characteristics.

Parameters	Raw water	Pretreated water
pH	7.8	7.9
Temp. (°C)	22.7	22.5
ORP (mV)	167	157
EC (mS/m)	80.5	79.7
Fe (II) (mg/L)	< 0.03	< 0.03
NH4-N (mg/L)	< 0.08	< 0.08
Alk. (mg/L CaCO3)	255	257
DOC (mg/L)	< 0.10	< 0.10
SiO2 (mg/L)	23.95	21.70
F (mg/L)	13.20	13.19

From the results in Table 4.4, it was found that the concentration of fluoride and silica in raw water was 13.20 and 23.95 mg/L. While the fluoride and silica concentration in pretreated water was 13.19 and 21.70 mg/L. The fluoride and silica concentration in raw water and pretreated water was almost the same. Then, it can be stated that pre-treatment process cannot remove fluoride and silica from raw water. The silica can be affected to the performance of groundwater defluoridation. So, the problem of this membrane plant is the membrane might be fouled with silica, which accumulation on membrane surface.

In December, 2008, groundwater from Pra Too Khong drinking water plant was collected. Fluoride concentration in groundwater was analysed, the results showed that fluoride concentration in Pra Too Khong drinking water plant groundwater was 11.99 – 15.17 mg/L, This value was higher than the standard of fluoride in drinking water. Then, groundwater from this site is recommended to filtrate by UTC-70 membrane before drinking.

In the groundwater defluoridation experiment, the permeate flux of each transmembrane pressure was calculated with the same equation in concentration polarization experimental. The result of permeate flux are shown in Figure 4.5.

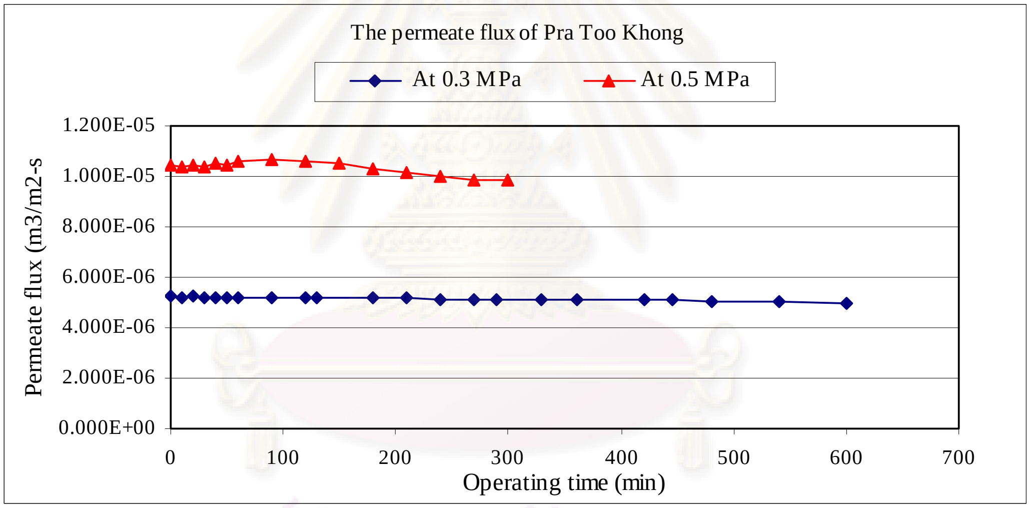


Figure 4.5 the permeate flux and operating time

The permeate flux at 1,000 ml permeate water obtained was 4.960×10^{-6} and 9.862×10^{-6} m^3/m^2-s under 0.3 and 0.5 MPa. The results showed that the permeate flux at 0.5 MPa was higher than permeate flux under 0.3 MPa. Furthermore, the operating time at 0.5 MPa was shorter than operating time under 0.3 MPa. From the results, it was found that the permeate flux from both transmembrane pressure were slightly decreased with the long time operating.

4.2.1.2 San Kam Pang hot spring well

The water characteristics of San Kam Pang hot spring well are reported in Table 4.5 (Wongrueng, 2008).

Table 4.5 San Kam Pang hot spring well groundwater characteristics

Parameters	San Kam Pang hot spring well
pH	8.9
EC (ms/cm)	76.1
Total Alk (mg/L as $CaCO_3$)	249
Na^+ (mg/L)	168.8
Ca^{2+} (mg/L)	<0.03
K^+ (mg/L)	12.0
SiO_2 (mg/L)	147.1
F^- (mg/L)	17.9
Cl^- (mg/L)	4.1
SO_4^{2-} (mg/L)	41.1

From Table 4.5, it was found that San Kam Pang hot spring well contains high fluoride concentration (17.9 mg/L) and silica concentration (147.1 mg/L).

In December, 2008, about 20 L of groundwater from San Kam Pang hot spring well was collected in plastic tank. The fluoride concentration was analysed and the result showed that groundwater from San Kam Pang hot spring well contained fluoride concentration 19.69 – 22.48 mg/L, which higher than the fluoride concentration of Pra Too Khong groundwater. The permeate flux was calculated and showed in Figure 4.6

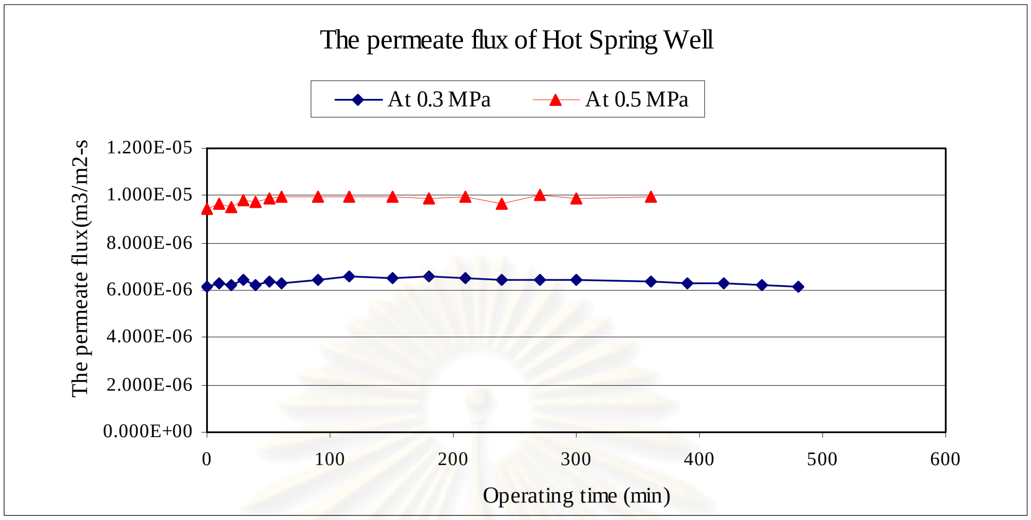


Figure 4.6 Permeate flux of San Kam Pang hot spring well

The permeate flux at 1,000 ml permeate water obtained was 6.127×10^{-6} and 9.921×10^{-6} m^3/m^2-s under 0.3 and 0.5 MPa. The results of permeate flux from San Kam Pang hot spring well was almost the same with permeate flux from Pra Too Khong drinking water plant. The higher permeate flux was obtained at 0.5 MPa and the shorter operating time (360 minutes) was obtained under 0.5 MPa.

If compared between the results of Pra Too Khong and San Kam Pang hot spring, it was found that the results of permeate flux from both sites were quite similar. The higher operating transmembrane pressure (0.5 MPa) was provided the higher permeate flux and shorter operating time. The permeate flux from both sites were quite similar due to the fluoride concentration in feed solution. Furthermore, it was found that the permeate flux was decreased with higher operating time due to the accumulation of fluoride concentration in feed tank.

This results related to the studied of Wongrueng (2006), which reported that the permeate water flux of membranes apparently increased with the operating transmembrane pressure.

Khankum, 2007 studied the effect of silica fouling on fluoride removal by ultra low pressure reverse osmosis membrane. The synthesis water with 15 mg/L fluoride concentration was used as feed solution. The permeate flux at 1,000 ml permeate obtained was 2.548 x 10^{-6}, 7.342 x 10^{-6} and 1.344 x 10^{-5} m^3/m^2-s at 0.1, 0.3 ahd 0.5 MPa was reported.

The comparisons between permeate flux from Pra Too Khong and San Kam Pang hot spring well and Khankum studied are shown in Table 4.6.

Table 4.6 The comparisons of permeate flux

Operating pressure	Pra Too Khong groundwater	San Kam Pang hot spring well	Synthesis water with 15 mg/L of fluoride (Khankum, 2007)
0.3	4.960x10^{-6}	6.127x10^{-6}	7.342 x 10^{-6}
0.5	9.862x10^{-6}	9.921x10^{-6}	1.344 x 10^{-5}

From the results, it was found that the permeate flux of synthesis water was higher than permeate flux of Pra Too Khong and San Kam Pang hot spring. Then, it can be indicated that the other parameters in groundwater including silica and sodium was affected to the permeate flux. If groundwater contains higher concentration of any parameter, the permeate flux was low. Water flux decreases with increasing feed solute concentration since the higher concentrations result in larger osmotic pressures and a smaller driving force across the membrane (Bhattacharyya et al., 1992 and Williams, M. E., 2003).

4.2.2 Membrane fouled layer

The membrane fouled layer was investigated. The fluoride concentration in bulk solution, permeate water was analysed and showed in Table 4.7 and 4.8, whereas, the fluoride concentration on gel layer surface was calculated from the following equation.

$$\left(J_v\right)_{solute} = k\ln\left(\frac{C_M - C_P}{C_B - C_P}\right) \qquad (4.5)$$

Where k = $\left(D/\delta\right)$= mass transfer coefficient, D = solute diffusion coefficient, δ = boundary layer thickness, $\left(J_v\right)_{solute}$ = permeate flux of solute solution, C_M = solute concentration prevailing on membrane surface, C_P = solute concentration in permeate solution, and C_B = solute concentration in bulk solution.

By substitute C_M with C_G and used the mass transfer coefficient (k) from the concentration polarization experiment (k = $2.73\text{x}10^{-5}$ and $1.75\text{x}10^{-5}$ m/s at 0.3 and 0.5 MPa, respectively). The fluoride concentration on gel layer surface was obtained and showed in Table 4.7 and 4.8.

Then, the fluoride concentration on membrane surface (C_M) was calculated from Equation (4-6).

$$J_i = k_i\left(C_M - C_P\right) \qquad (4.6)$$

Where J_i = solute flux (Jv $_{(solute)}$. C_p)

C_M = concentration of membrane surface

C_P = concentration of permeate

The average solute mass transfer coefficient (k_i) was used from the concentration polarization experiment (4.5 x 10^{-7} m/s at 0.3 and 0.5 MPa, respectively). Then the fluoride concentration on membrane surface was obtained and showed in Table 4.7 and 4.8.

Table 4.7 Fluoride concentrations in bulk solution, permeate water, gel layer surface and membrane surface (C_M) of Pra Too Khong groundwater.

Permeate obtained	TMP (Mpa)	*CP* (mg/L)	*CB* (mg/L)	*CM* (mg/L)	*CG* (mg/L)	*CM/CG* f'	%R
250 ml	0.3	0.55	13.99	7.81	16.80	0.46	96.1
500 ml		0.61	14.96	8.56	17.91	0.48	95.9
750 ml		0.68	15.91	9.49	19.02	0.50	95.7
1000 ml		0.70	17.63	9.56	21.00	0.46	96.0
250 ml	0.5	0.41	12.61	8.98	22.76	0.39	96.7
500 ml		0.48	13.20	10.38	23.66	0.44	96.4
750 ml		0.56	14.36	11.76	25.28	0.47	96.1
1000 ml		0.61	16.54	12.45	28.59	0.44	96.3

From the result in Table 4.7, it was found that fluoride concentration in permeate water was 0.55-0.70 and 0.41-0.61 mg/L under transmembrane pressure 0.3 and 0.5 MPa, which meet the standard of fluoride in drinking water of Ministry of Health (0.7 mg/L). However, trend of the fluoride concentration in permeate water was increased with time. Due to the condition of experiment that set as recycled mode, the fluoride concentration in feed tank was increased with time and then the high concentration in feed tank might be effect on the fluoride rejection rate.

Table 4.8 Fluoride concentrations in bulk solution, permeate water, gel layer surface and membrane surface (C_M) of San Kam Pang hot spring well.

Permeate obtained	TMP (Mpa)	*CP* (mg/L)	*CB* (mg/L)	*CM* (mg/L)	*CG* (mg/L)	*CM/CG* f'	%R
250 ml	0.3	0.73	21.91	12.88	27.64	0.47	96.7
500 ml		0.70	23.77	12.15	29.90	0.41	97.1
750 ml		0.73	25.84	12.48	32.38	0.39	97.2
1000 ml		0.77	28.01	12.74	33.10	0.38	97.3
250 ml	0.5	0.59	22.43	12.07	39.09	0.31	97.4
500 ml		0.62	24.12	12.80	42.19	0.30	97.4
750 ml		0.64	25.59	12.79	44.05	0.29	97.5
1000 ml		0.69	27.51	14.10	47.97	0.29	97.5

From Table 4.8, the result showed that the fluoride concentration in permeate water was 0.70-0.77 and 0.59-0.69 mg/L under transmembrane pressure 0.3 and 0.5 MPa. It was found that the fluoride concentration in permeate water was meet the standard only at 0.5 MPa transmembrane pressure. It was cause of high fluoride concentration in feed tank, which decreased the fluoride rejection rate. Then, it was indicated that the higher transmembrane was provide the higher quality of permeate water than lower transmembrane pressure.

The membrane fouled layer from both sites was investigated. From the result in Table 4.7 and 4.8, it was found that the fluoride concentration on gel layer surface (C_G) was higher than fluoride concentration on membrane surface (C_M) at any permeate volume obtained both in Pra Too Khong and San Kam Pang hot spring. From the profile of fluoride concentration that showed in Chapter 2, it was stated that the membrane fouled layer of both sites was fouled with polymerized fouled layer. When the polymerized fouled layer occurred, the fouled layer was form with highly dense gel layer. The dense gel layer can act as second filtration, which leaded the fluoride concentration on membrane surface was lower than on gel layer surface.

The *CM/CG* ratio in various times was investigated. The results from Table 4.7 and 4.8 showed that the *CM/CG* ratio at any permeates volumes were nearly constant. The average *CM/CG* ratio of Pra Too Khong groundwater was 0.42 and 0.49 under 0.3 and 0.5 MPa, respectively. While the average *CM/CG* ratio in hot spring was 0.36 and 0.34 at 0.3 and 0.5 MPa, respectively.

4.2.3 Fluoride rejection rate

When the fluoride concentration in bulk solution and permeate water was obtained. Then, the fluoride rejection rate was calculated.

From the results, it was found that the fluoride rejection rate from Pra Too Khong groundwater at any permeate volumes was nearly constant as shown in Table 4.7. The average of fluoride rejection rate was 95.9% and 96.4% under transmembrane pressure 0.3 and 0.5 MPa, respectively. For San Kam Pang hot spring well, the fluoride rejection rate at any permeate volumes was also nearly constant as shown in Table 4.8. The average of the fluoride rejection rate of was 97.1% and 97.5% at 0.3 and 0.5 MPa of transmembrane pressure.

These results correlated with the change of *CM/CG* ratio, when the *CM/CG* ratio decreased, the *CM* was decreased. Then, the fluoride rejection rate was goes up. Wongrueng (2007) reported that when the flux is constant during the course of filtration experiments, the fluoride rejection rates are determined by the changes of *CM/CG* ratio. If *CM/CG* ratio go down, then *CM* decreases and the fluoride rejection rate goes up.

From the results, it was found that the fluoride rejection rate at 0.3 and 0.5 MPa was quite similar. However, the higher fluoride rejection rate was obtained at higher transmembrane pressure. This may be due to a decrease in the average pore size on the membrane surface and increase in the preferential sorption of pure water at higher pressure, e.g., the solvent permeability increases compared to solute at high pressure (Ozaki et al., 2002).

Khankum, 2007 reported that the fluoride rejection rate of synthesis water with fluoride 15 mg/L was 95.5% and 95.1% under 0.3 and 0.5 MPa, respectively. If compared with this study, it was found that the fluoride rejection rate is almost same. Then, it can be indicated that the other parameters in groundwater not affect to the fluoride rejection rate in groundwater defluoridation by ULPRO membrane. This cause might be due to the low concentration of other parameter. In actually, another parameter such as caicium or magnesium can be affected on performance of groundwater defluoridation.

4.2.4 Effect of high silica concentration.

The effect of high silica concentration on groundwater defluoridation was investigated. High silica concentration was used to accelerate the fouling process. If used low concentration, we have to wait for several day. In real case, silica concentration in groundwater is low. But in some case of membrane plant, although silica is low, at high recovery rate the silica will high. In this experiment, the silica concentration 500 mg/L was added to groundwater from both sites.

4.2.4.1 Effect on permeate flux

The permeate flux of Pra Too Khong groundwater with silica added was calculated. It was found that the permeate flux at 1,000 ml permeate water obtained was 4.177×10^{-6} and 7.899×10^{-6} m^3/m^2-s under 0.3 and 0.5 MPa. The results of permeate flux with operating time at each transmembrane pressure was showed in Figure 4.7

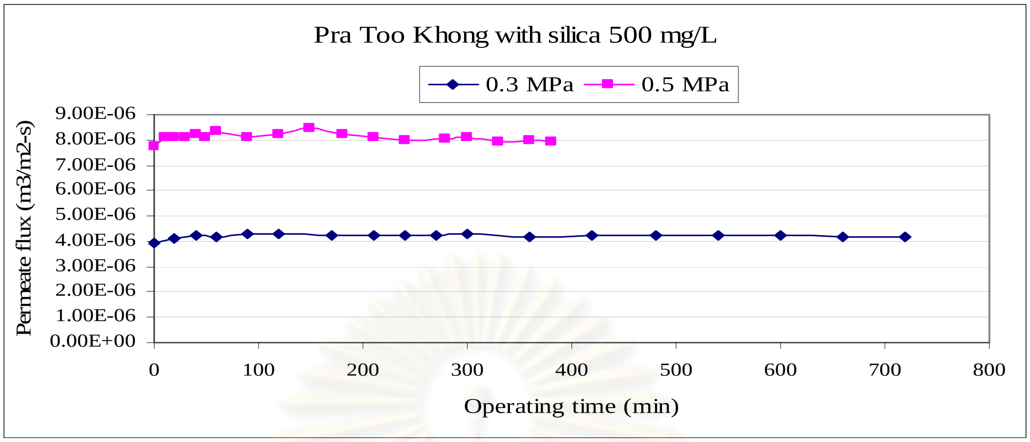


Figure 4.7 Permeate flux of Pra Too Khong groundwater with silica added

The permeate flux of San Kam Pang hot spring well with silica added was calculated. It was found that the permeate flux at 1,000 ml permeate water obtained was 4.296x10^{-6} and 7.153x10^{-6} m^3/m^2-s under 0.3 and 0.5 MPa. The results of permeate flux with operating time at each transmembrane pressure was showed in Figure 4.8.

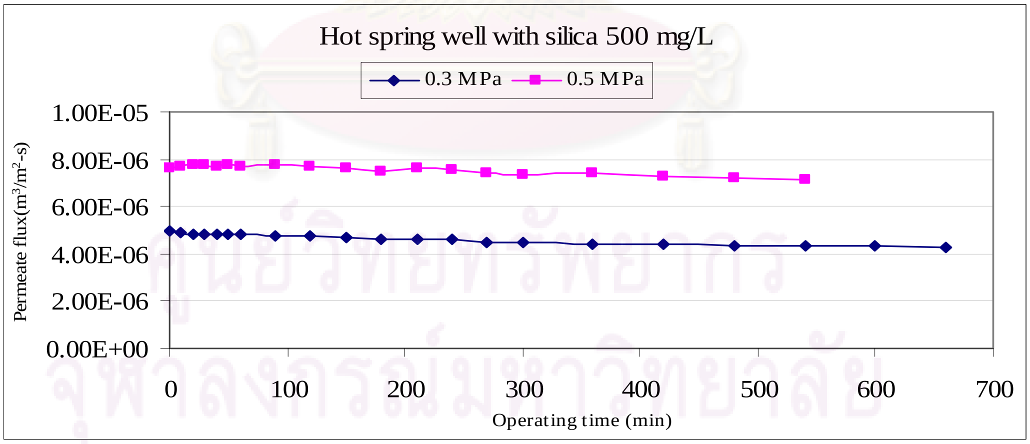


Figure 4.8 Permeate flux of groundwater from two sites with silica 500 mg/L

Khankum, 2007 studied the effect of silica fouling on fluoride removal by ultra low pressure reverse osmosis membrane. The synthesis water with 15 mg/L fluoride concentration was combined with 100 and 300 mg/L of silica concentrations used to prepare feed solution.

Table 4.9 illustrated the comparison of permeate flux between groundwater with and without silica and the permeate flux from Khankum studied.

Table 4.9 Permeate flux at various silica concentrations

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	Silica concentrations (mg/L)	Permeate flux (m^3/m^2-s)	
		0.3 MPa	0.5 MPa
Pra Too Khong groundwater	21.70^*	$4.960x10^{-6}$	$9.862x10^{-6}$
	521.70^{**}	$4.177x10^{-6}$	$7.899x10^{-6}$
San Kam Pang hot spring well	147.1^*	$6.127x10^{-6}$	$9.921x10^{-6}$
	647.1^{**}	$4.296x10^{-6}$	$7.153x10^{-6}$
Synthesis water with fluoride 15 mg/L (Khankum, 2007)	100	$7.278x10^{-6}$	$1.235x10^{-5}$
	300	$7.184x10^{-6}$	$1.217x10^{-5}$

(* Approximately silica concentration not added silica to groundwater, ** Approximately silica concentration after added silica 500 mg/L into groundwater)

From the results from Table. 4.9, it was found that the permeate flux from groundwater with added silica 500 mg/L was lower than permeate flux of groundwater without silica. In Pra Too Khong groundwater, the permeate flux was decrease from $4.960x10^{-6}$ to $4.177x10^{-6}$ and $9.862x10^{-6}$ to $7.899x10^{-6}$ m^3/m^2-s under 0.3 and 0.5 MPa. For San Kam Pang hot spring well, the permeate flux was decrease from $6.127x10^{-6}$ to $4.296x10^{-6}$ and $9.921x10^{-6}$ to $7.153x10^{-6}$ m^3/m^2-s under 0.3 and 0.5 MPa. From the result, it can be stated that the silica concentration in feed water was affected on permeate flux. The permeate flux was decreased with increasing of silica concentration in feed water. This may be attributed to the effect of the accumulation of silica in the feed water on the membrane surface (Liu et al., 1998). The effect of silica fouling was found to be the most influential on the flux declined (Matsui, 2007).

If compared with the results of Khankum studied, it was found that the permeate flux from both site without silica added was lower than the synthesis water with 100 and 300 mg/L of silica concentration. Then, it can be stated that some parameters in groundwater was affected to the permeate flux. The groundwater might be contains the higher concentration of any parameter than synthesis water with fluoride concentration 15 mg/L combined with 100 and 300 mg/L of silica concentration.

4.2.4.2 Silica fouled layer

The silica fouled layer in groundwater defluoridation by ULPRO membrane was investigated. The fluoride concentrations in bulk solutions, permeate water, gel layer surface and membrane surface was calculated with the same method in membrane fouled layer part. The results of both sites are showed separately in Table 4.10 and 4.11.

Table 4.10 Pra Too Khong groundwater with silica 500 mg/L

Permeate obtained	TMP (Mpa)	*CP* (mg/L)	*CB* (mg/L)	*CM* (mg/L)	*CG* (mg/L)	*CM/CG* f'	%R
250 ml	0.3	1.36	17.15	16.01	19.80	0.81	92.1
500 ml		1.41	18.21	16.44	20.99	0.78	92.3
750 ml		1.51	19.44	17.90	22.45	0.80	92.2
1000 ml		1.64	20.78	19.14	23.95	0.80	92.1
250 ml	0.5	0.75	13.81	12.68	21.49	0.59	94.6
500 ml		0.76	14.41	13.18	22.63	0.58	94.7
750 ml		0.87	16.53	14.60	25.68	0.57	94.8
1000 ml		0.99	16.71	16.34	25.69	0.64	94.1

Table 4.11 San Kam Pang hot spring well with silica 500 mg/L

Permeate obtained	TMP (Mpa)	*CP* (mg/L)	*CB* (mg/L)	*CM* (mg/L)	*CG* (mg/L)	*CM/CG* f'	%R
250 ml	0.3	1.67	20.73	21.59	24.29	0.89	91.9
500 ml		1.74	21.94	21.57	25.53	0.84	92.1
750 ml		1.82	23.24	22.06	26.94	0.82	92.2
1000 ml		1.99	25.15	23.80	29.10	0.82	92.1
250 ml	0.5	1.53	22.86	24.86	34.74	0.72	93.3
500 ml		1.65	24.56	25.99	36.84	0.71	93.3
750 ml		1.79	26.09	27.68	38.75	0.71	93.1
1000 ml		1.87	27.91	28.20	41.05	0.69	93.3

The results from table 4.10 and 4.11 showed that the fluoride concentration on gel layer surface was higher than fluoride concentration on membrane surface. Then, it can be indicated that membrane was fouled with polymerized fouled layer (dense-gel layer) at all case. Matsui, 2007 reported that SiO_2 was saturated on the membrane surface. The gel layer formed by monosilicic silica was found to be densely polymerized and low permeability. It was also found that the polymerization of silica was increased with increasing of initial silica concentration (R. Sheikholeslami et al, 2001). At silica concentrations greater than 300 ppm, polymerization took place even in the absence of calcium and magnesium (Sheikholeslami et al, 2001). Furthermore, the fluoride concentration in permeate water were higher than fluoride concentration in permeate water of groundwater (not added silica). So, it can be stated that silica concentration in feed water might be affect on fluoride rejection rate. The higher silica concentration in feed water was provided the higher fluoride concentration in permeate water.

Khankum, 2007 reported that at 0.3 and 0.5 MPa, the formation of a polymerized silica fouled layer occurred at a low silica concentration of 100 mg/L, whereas the formation of colloidal silica fouled layer appeared at a high silica concentration of 300 mg/L. From the results of my studied, it was found that the polymerized fouled layer was occurred when 500 mg/L silica added. This result might be due to the induction time of silica.

The *CM/CG* ratio at various times was investigated. From Table 4.10 and 4.11, it was found that the *CM/CG* ratio was almost the same. The average of *CM/CG* ratio in Pra Too Khong groundwater was 0.70 and 0.67 at 0.3 and 0.5 MPa, respectively. Whereas, in hot spring, the average of *CM/CG* ratio was 0.74 and 0.79 under 0.3 and 0.5 MPa, respectively.

The results of this part were compared with the results of groundwater without silica added. From the comparisons, it was found that when adding silica to groundwater, the *CM/CG* ratio was increased. In Pra Too Khong groundwater, the *CM/CG* ratio was increase from 0.40 to 0.71 and 0.49 to 0.71 at 0.3 and 0.5 MPa, respectively. While, in hot spring, the *CM/CG* ratio was increase from 0.34 to 0.72 and 0.33 to 0.77 under 0.3 and 0.5 MPa, respectively. From the results of CG, it was found that the *CG* was nearly constant. Then, when *CM/CG* was increased, it means that the *CM* was increased. From the results, it can be indicated that when silica was added, silica might be affect on the layer of membrane surface. It makes the layer more thickness than the experiment without silica added. When the layer was more thickness, the back diffusion of fluoride was difficult. So, the higher fluoride concentration was accumulated on the membrane surface. Then, the fluoride concentration on membrane surface was high.

4.2.4.3 Fluoride rejection rate

The fluoride rejection rate when adding silica concentration 500 mg/L into groundwater was investigated. It was found that the fluoride rejection rate at any permeate volumes was almost the same as shown in Table 4.10 and 4.11. The average of fluoride rejection rate of Pra Too Khong groundwater with silica 500 mg/L was 92.2% and 94.6% at transmembrane pressure 0.3 and 0.5 MPa, respectively. Whereas, the average of fluoride rejection rate of San Kam Pang hot spring well with silica concentration 500 mg/L was 92.1% and 93.3% at 0.3 and 0.5 MPa, respectively.

From the results of fluoride rejection rate, it was found that the higher transmembrane pressure was provided the higher rejection rate.

If compared between the groundwater with silica adding and without silica adding, it was found that the fluoride rejection rate of groundwater with silica adding was lower than in groundwater not adding silica at all transmembrane pressure. In Pra Too Khong groundwater, the fluoride rejection rate was decrease from 96.0% to 92.1% and 96.3% to 94.1% under 0.3 and 0.5 MPa, respectively, when silica was added. While, in hot spring groundwater, when added silica, the fluoride rejection rate was decrease from 97.3% to 92.1% and 97.5% to 93.3% at 0.3 and 0.5 MPa, respectively. The fluoride rejection rate of groundwater with silica added was decreased due to the increasing of the *CM/CG* ratio.

From the results, it can be indicated that the silica concentration was affected on fluoride rejection rate. The fluoride rejection rate was decreased with the increasing of concentration in feed water. The increase in salt concentration at the membrane surface created greater salt concentration gradient across the membrane, causing an increase of salt concentration in permeate and a decrease in salt rejection by the RO membrane (How, 2005).

Khankum, 2007 reported that the percentages of fluoride rejection of synthesis water with fluoride 15 mg/L combined with 100 mg/L of silica was 98.7% and 98.8% at 0.3 and 0.5 MPa. For the synthesis water with fluoride 15 mg/L combined with 300 mg/L of silica, the fluoride rejection rate was 94.9% and 98.0% under 0.3 and 0.5 MPa.

The results of fluoride rejection rate of groundwater with and without silica were compared with the fluoride rejection rate of Khankum studied and shown in Table 4.12.

Table 4.12. The comparison of fluoride rejection rate

	Silica concentrations (mg/L)	Fluoride rejection rate (%)	
		0.3 MPa	0.5 MPa
Pra Too Khong groundwater	21.70*	95.9	96.4
	521.70**	92.2	94.6
San Kam Pang hot spring well	147.1*	97.1	97.5
	647.1**	92.1	93.3
Synthesis water with fluoride 15 mg/L (Khankum, 2007)	0	95.5	95.1
	100	98.7	98.8
	300	94.9	98.0

(* Approximately silica concentration not added silica to groundwater, ** Approximately silica concentration after added silica 500 mg/L into groundwater)

From the results, it was found that the fluoride rejection rate was decreased with the increasing of silica concentration in feed solution. When add 500 mg/L of silica into groundwater, the fluoride rejection rate was decreased in both sites. From studied of Khankum (2007), it was reported that fluoride rejection rate at 100 mg/L of silica added was higher than in without silica added. This cause of the polymerized silica fouled layer occurred on membrane surface and acts as second filtration. So, the fluoride rejection rate was increased. In case of 500 mg/L of silica added, the membrane was fouled with polymerized fouled layer but the fluoride rejection rate was lower. It might be related with the thickness of membrane fouled layer. If the thickness of membrane fouled layer increased, the back diffusion of fluoride is difficulty. Then, the fluoride rejection rate was decreased. From the results, it can be indicated that the silica might be affected on the membrane layer by increase the thickness of membrane fouled layer, which leading to decrease in fluoride ejection rate.

CHAPTER V

CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

The results from silica fouling of UTC-70 ultra low pressure reverse osmosis membrane in defluoridation of Chiang mai basin groundwater can be concluded as follows:

1. In the concentration polarization experiment, the mass transfer coefficient in solution was estimated. It was found that the average mass transfer coefficient in solution were $2.73\text{x}10^{-5}$ and $1.75\text{x}10^{-5}$ m/s at transmembrane pressure 0.3 and 0.5 MPa, respectively. Furthermore, the solute mass transfer coefficient through membrane was estimated. The result showed that the solute mass transfer coefficient through membrane was $3.92\text{x}10^{-7}$ and $5.08\text{x}10^{-7}$ m/s at 0.3 and 0.5 MPa, respectively.

2. The permeate flux of each experiment were investigated. In the concentration polarization experiment, the permeate flux was in range $7.899\text{x}10^{-6}$- $2.012\text{x}10^{-6}$ and $1.41\text{x}10^{-5}$- $4.188\text{x}10^{-6}$ m^3/m^2-s at 0.3 and 0.5 MPa, respectively. The permeate flux was decreased with increasing of fluoride concentration in feed water. In groundwater defluoridation, the permeate flux of Pra Too Khong groundwater was in range $6.13\text{x}10^{-6}$ - $6.56\text{x}10^{-6}$ and $9.69\text{x}10^{-6}$ - $9.98\text{x}10^{-6}$ m3/m2-s at 0.3 and 0.5 MPa, respectively, whereas, the permeate flux of San Kam Pang hot spring well was $4.96\text{x}10^{-6}$ – $5.18\text{x}10^{-6}$ and $9.86\text{x}10^{-6}$ – $1.06\text{x}10^{-5}$ m3/m2-s at 0.3 and 0.5 MPa, respectively.

3. The fluoride concentration in groundwater from Pra Too Khong and hot spring was investigated. The result showed that the fluoride concentration in Pra Too

Khong groundwater was 11.99 – 15.17 mg/L, whereas, in hot spring was found to contain fluoride concentration 19.69 – 22.48 mg/L. When groundwater from both sites was filtrated with UTC-70 ULPRO membranes, the fluoride concentration in permeate water was 0.41- 0.70 mg/L and 0.59 – 0.77 mg/L at Pra Too Khong and hot spring, respectively.

4. The fluoride rejection rate was investigated. It was found that the average fluoride rejection rate of Pra Too Khong groundwater was 95.9% and 96.4% at transmembrane pressure 0.3 and 0.5 MPa, respectively, whereas, the average fluoride rejection rate of San Kam Pang hot spring well was 97.1% and 97.5% at 0.3 and 0.5 MPa, respectively. The higher fluoride rejection rate was obtained at higher operating transmembrane pressure. The comparison between two sites, it was found that the fluoride rejection rate in Pra too Khong groundwater was lower than fluoride rejection rate in hot spring under transmembrane pressure 0.3 and 0.5 MPa

5. The membrane fouling was investigated. The fluoride concentration on membrane surface and on gel layer was calculated. The result showed that the fluoride concentration on gel layer surface was higher than fluoride concentration on membrane surface. Then, it was indicated that the polymerized fouled layer was occurred both in Pra Too Khong groundwater and San Kam Pang hot spring well.

6. The effect of silica on groundwater defluoridation was investigated. It was found that silica was effect on permeate flux and fluoride rejection rate. The permeate flux and fluoride rejection rate decreased with increasing of silica concentration in feed solution. The average of fluoride rejection rate of Pra Too Khong was 92.2% and 94.6% under 0.3 and 0.5 MPa, whereas, in hot spring, the average fluoride rejection rate was 92.1% and 93.3% at 0.3 and 0.5 MPa.

7. The silica fouling on membrane was investigated by measured the fluoride concentration on gel layer and on membrane surface. The result showed that the fluoride concentration on gel layer was higher than fluoride concentration on membrane surface at all case. It can be indicated that membrane was fouled with

polymerized silica fouled layer. Furthermore, it was found that silica can increase the thickness of membrane surface layer, which leading to increase concentration on membrane surface and decrease in fluoride rejection rate.

5.2 Recommendations for future research

Based on the results of this study, the recommendation for future studies can be proposed.

1. In this study the effect of silica on groundwater defluoridation in the short-term operating (until 1,000 ml of permeate water obtained) was investigated. Then, the effect of silica in the long-term operating should be investigated such as 1 or 2 day operating.

2. Based on the results, it is important to remove silica before filtrated with UTC-70 membrane. Then, the pretreatment to remove silica are recommended to be studied.

3. The chemical cleaning method for membrane fouling is recommended to be investigated. There are many methods to solving the membrane fouling such as oxidants and acidic agents. Then, the future research to find the most effective method to cleaning the membrane fouling are recommended to be investigated.

4. The effect of some parameters including pH, temperature and presence of other ions on the formation of silica on membrane surface should be investigated.

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APPENDICES

APPENDIX A

CONCENTRATION POLARIZATION EXPERIMENT: PERMEATE FLUX

Table A-1 Permeate flux until 250 ml of permeate water obtained at fluoride concentration 0 mM at 0.3 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m^3/m^2.sec)
0	1.40	0.307	0.292	211	7.899E-06
10	1.40	0.307	0.292	211	7.899E-06
20	1.40	0.308	0.293	214	7.788E-06
30	1.40	0.308	0.292	208	8.013E-06
40	1.40	0.308	0.292	215	7.752E-06
50	1.40	0.308	0.292	212	7.862E-06
60	1.40	0.309	0.293	211	7.899E-06
90	1.40	0.308	0.293	213	7.825E-06
120	1.40	0.308	0.293	210	7.937E-06
150	1.40	0.308	0.293	211	7.899E-06

Table A-2 Permeate flux until 250 ml of permeate water obtained at fluoride concentration 10 mM at 0.3 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	1.32	0.306	0.293	275	6.061E-06
10	1.32	0.307	0.293	277	6.017E-06
20	1.32	0.307	0.293	273	6.105E-06
30	1.32	0.306	0.294	273	6.105E-06
40	1.32	0.306	0.292	277	6.017E-06
50	1.32	0.306	0.292	272	6.127E-06
60	1.32	0.306	0.292	274	6.083E-06
90	1.32	0.307	0.293	274	6.083E-06
120	1.32	0.306	0.293	273	6.105E-06
150	1.32	0.306	0.294	275	6.061E-06

Table A-3 Permeate flux until 250 ml of permeate water obtained at fluoride concentration 25 mM at 0.3 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	1.32	0.306	0.293	396	4.209E-06
15	1.32	0.306	0.293	392	4.252E-06
30	1.32	0.306	0.293	392	4.252E-06
45	1.32	0.305	0.292	391	4.263E-06
60	1.32	0.306	0.293	389	4.284E-06
90	1.32	0.307	0.293	390	4.274E-06
120	1.32	0.306	0.293	392	4.252E-06
150	1.32	0.306	0.293	392	4.252E-06
180	1.32	0.306	0.293	392	4.252E-06
195	1.32	0.307	0.292	392	4.252E-06

Table A-4 Permeate flux until 250 ml of permeate water obtained at fluoride concentration 50 mM at 0.3 MPa

Time (min)	Flow l/min	pressure in Mpa	pressure out Mpa	Time at V=10 ml (sec)	Flux (m3/m2.sec)
0	1.32	0.305	0.291	813	2.050E-06
20	1.32	0.306	0.292	813	2.050E-06
40	1.32	0.305	0.291	815	2.045E-06
60	1.32	0.307	0.292	811	2.055E-06
90	1.32	0.307	0.293	812	2.053E-06
120	1.32	0.307	0.293	816	2.042E-06
150	1.32	0.307	0.293	820	2.033E-06
180	1.32	0.307	0.293	819	2.035E-06
210	1.32	0.307	0.293	823	2.025E-06
240	1.32	0.307	0.293	822	2.028E-06
270	1.32	0.307	0.293	824	2.023E-06
300	1.32	0.307	0.293	825	2.020E-06
330	1.32	0.306	0.293	828	2.013E-06

Table A-5 Permeate flux until 250 ml of permeate water obtained at fluoride concentration 0 mM at 0.5 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	0.08	0.486	0.485	117	1.425E-05
10	0.08	0.486	0.485	113	1.475E-05
20	0.08	0.483	0.482	118	1.412E-05
30	0.08	0.482	0.481	119	1.401E-05
40	0.08	0.483	0.481	116	1.437E-05
50	0.08	0.483	0.481	115	1.449E-05
60	0.12	0.483	0.482	115	1.449E-05
75	0.08	0.484	0.482	118	1.412E-05

Table A-6 Permeate flux until 250 ml of permeate water obtained at fluoride concentration 10 mM at 0.5 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	0.20	0.484	0.482	145	1.149E-05
10	0.20	0.486	0.484	143	1.166E-05
20	0.20	0.485	0.483	142	1.174E-05
30	0.20	0.487	0.486	142	1.174E-05
40	0.20	0.485	0.483	142	1.174E-05
50	0.20	0.484	0.482	143	1.166E-05
60	0.20	0.484	0.482	144	1.157E-05
80	0.24	0.485	0.482	144	1.157E-05

Table A-7 Permeate flux until 250 ml of permeate water obtained at fluoride concentration 25 mM at 0.5 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	0.16	0.480	0.479	183	9.107E-06
10	0.08	0.478	0.477	180	9.259E-06
20	0.12	0.473	0.471	179	9.311E-06
30	0.16	0.467	0.466	181	9.208E-06
40	0.12	0.476	0.475	182	9.158E-06
50	0.12	0.476	0.475	185	9.009E-06
60	0.08	0.476	0.474	183	9.107E-06
90	0.12	0.470	0.468	189	8.818E-06

Table A-8 Permeate flux until 250 ml of permeate water obtained at fluoride concentration 50 mM at 0.5 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	0.12	0.471	0.470	384	4.340E-06
10	0.12	0.473	0.472	386	4.318E-06
20	0.12	0.474	0.473	384	4.340E-06
30	0.12	0.473	0.472	385	4.329E-06
40	0.12	0.472	0.471	387	4.307E-06
50	0.12	0.471	0.470	385	4.329E-06
60	0.12	0.472	0.471	388	4.296E-06
90	0.12	0.471	0.470	392	4.252E-06
120	0.12	0.471	0.471	390	4.274E-06
160	0.12	0.471	0.470	398	4.188E-06

APPENDIX B

PRA TOO KHONG GROUNDWATER DEFLUORIDATION

Table B-1 Permeate flux of Pra Too Khong groundwater until 1,000 ml of permeate water obtained at 0.3 MPa

Time (min)	Flow l/min	pressure in Mpa	pressure out Mpa	Time at V=10 ml (sec)	Flux (m3/m2.sec)
0	1.16	0.306	0.292	319	5.225E-06
10	1.28	0.306	0.292	321	5.192E-06
20	1.28	0.306	0.292	319	5.225E-06
30	1.28	0.307	0.293	320	5.208E-06
40	1.28	0.306	0.293	321	5.192E-06
50	1.28	0.306	0.293	323	5.160E-06
60	1.28	0.306	0.293	321	5.192E-06
90	1.28	0.306	0.293	323	5.160E-06
120	1.28	0.307	0.293	322	5.176E-06
130	1.28	0.306	0.293	322	5.176E-06
180	1.28	0.306	0.293	323	5.160E-06
210	1.28	0.307	0.293	323	5.160E-06
240	1.28	0.306	0.293	324	5.144E-06
270	1.28	0.307	0.293	324	5.144E-06
289	1.28	0.309	0.293	326	5.112E-06
330	1.28	0.307	0.293	324	5.144E-06
360	1.28	0.308	0.294	326	5.112E-06
420	1.28	0.308	0.294	326	5.112E-06
445	1.28	0.306	0.293	328	5.081E-06
480	1.28	0.305	0.293	329	5.066E-06
540	1.28	0.306	0.293	332	5.020E-06
600	1.28	0.306	0.293	336	4.960E-06

Table B-2 Permeate flux of Pra Too Khong until 1,000 ml of permeate water obtained at 0.5 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	0.20	0.481	0.48	160	1.042E-05
10	0.20	0.476	0.475	161	1.035E-05
20	0.24	0.474	0.473	160	1.042E-05
30	0.28	0.475	0.474	161	1.035E-05
40	0.28	0.476	0.474	158	1.055E-05
50	0.28	0.475	0.473	160	1.042E-05
60	0.32	0.475	0.473	157	1.062E-05
90	0.28	0.481	0.479	156	1.068E-05
120	0.28	0.481	0.481	157	1.062E-05
150	0.28	0.482	0.480	159	1.048E-05
180	0.32	0.478	0.476	162	1.029E-05
210	0.32	0.475	0.473	164	1.016E-05
240	0.32	0.475	0.473	167	9.980E-06
270	0.28	0.475	0.473	169	9.862E-06
300	0.28	0.475	0.473	169	9.862E-06

Table B-3 pH and electro conductivity (EC) at 0.3 MPa

	Permeate volume obtained	pH	EC (μs/cm)
Feed		8.37	834
Concentrated water	250 ml	8.61	872
	500 ml	8.54	924
	750 ml	8.79	982
	1000 ml	9.16	1058
Permeate water	250 ml	8.73	76.7
	500 ml	9.00	81.5
	750 ml	9.11	86.4
	1000 ml	9.23	102.0

Table B-4 pH and electro conductivity (EC) at 0.5 MPa

	Permeate volume obtained	pH	EC (μs/cm)
Feed		8.23	737
Concentrated water	250 ml	8.51	764
	500 ml	8.58	808
	750 ml	8.67	864
	1000 ml	8.95	921
Permeate water	250 ml	8.45	57.4
	500 ml	8.75	58.0
	750 ml	8.93	59.6
	1000 ml	9.12	63.1

APPENDIX C

PRA TOO KHONG GROUNDWATER WITH SILICA CONCENTRATION 500 mg/L

Table C-1 Permeate flux of Pra Too Khong groundwater with silica 500 mg/L until 1,000 ml of permeate water obtained at 0.3 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	1.28	0.307	0.294	421	3.959E-06
20	1.28	0.305	0.292	407	4.095E-06
40	1.28	0.307	0.295	396	4.209E-06
60	1.28	0.305	0.292	397	4.198E-06
90	1.28	0.304	0.294	389	4.284E-06
120	1.28	0.304	0.294	388	4.296E-06
170	1.28	0.305	0.292	393	4.241E-06
210	1.28	0.304	0.291	394	4.230E-06
240	1.28	0.304	0.290	395	4.219E-06
270	1.28	0.304	0.290	393	4.241E-06
300	1.28	0.307	0.295	391	4.263E-06
360	1.28	0.307	0.295	399	4.177E-06
420	1.28	0.305	0.292	393	4.241E-06
480	1.28	0.308	0.295	394	4.230E-06
540	1.28	0.307	0.294	393	4.241E-06
600	1.28	0.308	0.295	392	4.252E-06
660	1.28	0.308	0.295	399	4.177E-06
720	1.28	0.305	0.292	399	4.177E-06

Table C-2 Permeate flux of Pra Too Khong groundwater with silica 500 mg/L until 1,000 ml of permeate water obtained at 0.5 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	0.16	0.462	0.462	215	7.752E-06
10	0.16	0.462	0.461	205	8.130E-06
20	0.16	0.463	0.462	206	8.091E-06
30	0.16	0.461	0.461	205	8.130E-06
40	0.16	0.461	0.460	203	8.210E-06
50	0.16	0.461	0.460	205	8.130E-06
60	0.16	0.461	0.460	200	8.333E-06
90	0.16	0.459	0.458	206	8.091E-06
120	0.12	0.461	0.460	203	8.210E-06
150	0.12	0.461	0.460	197	8.460E-06
180	0.12	0.460	0.459	202	8.251E-06
210	0.12	0.456	0.455	205	8.130E-06
240	0.12	0.459	0.457	208	8.013E-06
279	0.12	0.459	0.458	207	8.052E-06
300	0.12	0.460	0.459	205	8.130E-06
330	0.12	0.460	0.459	211	7.899E-06
360	0.08	0.460	0.459	208	8.013E-06
380	0.08	0.461	0.460	211	7.899E-06

Table C-3 pH and electro conductivity (EC) at 0.3 MPa

	Permeate volume obtained	pH	EC (μs/cm)
Feed		10.30	1986
Concentrated water	250 ml	10.27	2070
	500 ml	10.22	2170
	750 ml	10.07	2300
	1000 ml	10.05	2450
Permeate water	250 ml	10.18	232
	500 ml	10.20	236
	750 ml	10.04	234
	1000 ml	10.14	246

Table C-4 pH and electro conductivity (EC) at 0.5 MPa

	Permeate volume obtained	pH	EC (μs/cm)
Feed		10.11	1959
Concentrated water	250 ml	10.15	2030
	500 ml	10.19	2120
	750 ml	10.24	2250
	1000 ml	10.18	2380
Permeate water	250 ml	10.07	204.0
	500 ml	10.15	194.3
	750 ml	10.12	189.4
	1000 ml	10.08	200.0

APPENDIX D

HOT SPRING GROUNDWATER DEFLUORIDATION

Table D-1 Permeate flux of San Kam Pang hot spring well until 1,000 ml of permeate water obtained at 0.3 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	1.56	0.31	0.291	271	6.150E-06
10	1.56	0.308	0.290	264	6.313E-06
20	1.56	0.308	0.290	268	6.219E-06
30	1.56	0.308	0.290	259	6.435E-06
40	1.56	0.308	0.290	267	6.242E-06
50	1.56	0.308	0.290	261	6.386E-06
60	1.56	0.308	0.290	265	6.289E-06
90	1.56	0.308	0.291	258	6.460E-06
115	1.56	0.308	0.290	254	6.562E-06
150	1.56	0.311	0.290	256	6.510E-06
180	1.56	0.309	0.290	254	6.562E-06
210	1.56	0.309	0.290	257	6.485E-06
240	1.56	0.309	0.290	258	6.460E-06
270	1.56	0.309	0.290	259	6.435E-06
300	1.56	0.310	0.291	258	6.460E-06
360	1.56	0.310	0.291	263	6.337E-06
390	1.56	0.308	0.289	266	6.266E-06
420	1.56	0.308	0.289	265	6.289E-06
450	1.56	0.309	0.291	267	6.242E-06
480	1.56	0.309	0.291	272	6.127E-06

Table D-2 Permeate flux of San Kam Pang hot spring well until 1,000 ml of permeate water obtained at 0.5 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	0.64	0.493	0.485	177	9.416E-06
10	0.68	0.504	0.498	173	9.634E-06
20	0.72	0.502	0.494	175	9.524E-06
30	0.80	0.504	0.496	170	9.804E-06
40	0.80	0.504	0.496	171	9.747E-06
50	0.84	0.502	0.494	169	9.862E-06
60	0.84	0.504	0.496	168	9.921E-06
90	0.84	0.504	0.496	167	9.980E-06
115	0.84	0.504	0.496	167	9.980E-06
150	0.84	0.504	0.496	167	9.980E-06
180	0.84	0.503	0.495	169	9.862E-06
210	0.84	0.504	0.496	167	9.980E-06
240	0.84	0.504	0.496	172	9.690E-06
270	0.84	0.502	0.495	166	1.004E-05
300	0.84	0.502	0.495	169	9.862E-06
360	0.84	0.502	0.495	168	9.921E-06

Table D-3 pH and electro conductivity (EC) at 0.3 MPa

	Permeate volume obtained	pH	EC (μs/cm)
Feed		8.26	759
Concentrated water	250 ml	8.33	797
	500 ml	8.47	853
	750 ml	8.54	919
	1000 ml	8.67	993
Permeate water	250 ml	8.57	42.0
	500 ml	8.77	46.7
	750 ml	9.02	51.2
	1000 ml	9.22	57.1

Table D-4 pH and electro conductivity (EC) at 0.5 MPa

	Permeate volume obtained	pH	EC (μs/cm)
Feed		8.15	759
Concentrated water	250 ml	8.21	793
	500 ml	8.32	845
	750 ml	8.42	903
	1000 ml	8.51	969
Permeate water	250 ml	8.23	37.8
	500 ml	8.35	38.4
	750 ml	8.56	43.4
	1000 ml	8.81	47.6

APPENDIX E

HOT SPRING GROUNDWATER WITH SILICA CONCENTRATION 500 mg/L

Table E-1 Permeate flux of San Kam Pang hot spring well with silica 500 mg/L until 1,000 ml of permeate water obtained at 0.3 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	1.16	0.305	0.292	335	4.975E-06
10	1.24	0.305	0.293	341	4.888E-06
20	1.24	0.306	0.294	346	4.817E-06
30	1.24	0.304	0.291	344	4.845E-06
40	1.24	0.307	0.295	346	4.817E-06
50	1.24	0.307	0.295	343	4.859E-06
60	1.24	0.307	0.295	345	4.831E-06
90	1.24	0.304	0.293	349	4.776E-06
120	1.24	0.307	0.294	353	4.721E-06
150	1.24	0.305	0.292	357	4.669E-06
180	1.24	0.306	0.294	363	4.591E-06
210	1.24	0.306	0.294	363	4.591E-06
240	1.28	0.307	0.295	363	4.591E-06
270	1.28	0.306	0.294	373	4.468E-06
300	1.28	0.306	0.295	373	4.468E-06
360	1.28	0.308	0.296	377	4.421E-06
420	1.28	0.306	0.294	380	4.386E-06
480	1.28	0.306	0.294	383	4.352E-06
540	1.28	0.306	0.294	387	4.307E-06
600	1.28	0.306	0.294	386	4.318E-06
660	1.28	0.306	0.294	388	4.296E-06

Table E-2 Permeate flux of San Kam Pang hot spring well with silica 500 mg/L until 1,000 ml of permeate water obtained at 0.5 MPa

Time	Flow	pressure in	pressure out	Time at V=10 ml	Flux
(min)	l/min	Mpa	Mpa	(sec)	(m3/m2.sec)
0	0.24	0.470	0.469	218	7.645E-06
10	0.24	0.471	0.469	216	7.716E-06
20	0.24	0.471	0.469	214	7.788E-06
30	0.24	0.470	0.469	215	7.752E-06
40	0.24	0.469	0.467	216	7.716E-06
50	0.24	0.469	0.468	214	7.788E-06
60	0.24	0.469	0.467	217	7.680E-06
90	0.24	0.470	0.468	215	7.752E-06
120	0.20	0.469	0.467	216	7.716E-06
150	0.20	0.468	0.467	218	7.645E-06
180	0.20	0.469	0.468	222	7.508E-06
210	0.24	0.470	0.469	218	7.645E-06
240	0.24	0.472	0.470	221	7.541E-06
270	0.24	0.472	0.470	224	7.440E-06
300	0.24	0.472	0.469	227	7.342E-06
360	0.24	0.472	0.469	224	7.440E-06
420	0.24	0.472	0.469	230	7.246E-06
480	0.24	0.472	0.469	231	7.215E-06
540	0.24	0.472	0.469	233	7.153E-06

Table E-3 pH and electro conductivity (EC) at 0.3 MPa

	Permeate volume obtained	pH	EC (μs/cm)
Feed		10.52	1973
Concentrated water	250 ml	10.47	2050
	500 ml	10.41	2150
	750 ml	10.35	2250
	1000 ml	10.28	2390
Permeate water	250 ml	10.11	248
	500 ml	10.09	255
	750 ml	10.12	270
	1000 ml	10.04	265

Table E-4 pH and electro conductivity (EC) at 0.5 MPa

	Permeate volume obtained	pH	EC (ms/cm)
Feed		10.63	1978
Concentrated water	250 ml	10.61	2060
	500 ml	10.63	2180
	750 ml	10.56	2310
	1000 ml	10.56	2450
Permeate water	250 ml	10.24	245
	500 ml	10.31	234
	750 ml	10.24	225
	1000 ml	10.33	232

APPENDIX F

THE MASS TRANSFER COEFFICIENT IN SOLUTION (k)

Mass transfer coefficient (k) was calculated by using the equation (2-7)

$$k = \frac{\left(J_v\right)_{solute}}{\ln\left\{\dfrac{\Delta P}{\pi_B - \pi_P} \cdot \left[1 - \dfrac{\left(J_v\right)_{solute}}{\left(J_v\right)_{H_2O}}\right]\right\}}$$

Where $\left(J_v\right)_{H_2\mathrm{O}}$ = permeate flux of pure water, ΔP = transmembrane pressure, π_B = osmotic pressure of bulk solution, and π_P = osmotic pressure of permeate solution.

Example: at 0.3 MPa

Run no.	Feed solution	Transmembrane pressure (Mpa)	Concentrated water (C_B)		Permeate water (C_P)		$\Delta\pi$	Permeate flux (m^3/m^2-s)
			mM	π_B	mM	π_P	$\pi_B-\pi_P$	Jv x 10^{-5}
A1	0	0.3	0	0	0	0	0	0.789
A2	9.80		11.81	0.0545	0.60	0.0028	0.0518	0.606
A3	24.21		28.58	0.1320	2.83	0.0131	0.1189	0.425
A4	49.81		55.24	0.2551	9.74	0.0450	0.2101	0.201

Run no. A2:

$$k = \frac{6.061x10^{-6}}{\ln\left\{\dfrac{0.300}{0.0545 - 0.0028} \cdot \left[1 - \dfrac{6.061x10^{-6}}{7.899x10^{-6}}\right]\right\}}$$

$k = 2.021\text{x}10^{-5}$ m/s

Run no. A3: $$k = \frac{4.253x10^{-6}}{\ln\left\{\frac{0.299}{0.1320-0.0131}\cdot\left[1-\frac{4.253x10^{-6}}{7.899x10^{-6}}\right]\right\}}$$

k = 2.823x10^{-5} m/s

Run no. A4: $$k = \frac{2.012x10^{-6}}{\ln\left\{\frac{0.299}{0.2551-0.0450}\cdot\left[1-\frac{2.012x10^{-6}}{7.899x10^{-6}}\right]\right\}}$$

k = 3.334x10^{-5} m/s

Table D-1 mass transfer coefficient at 0.3 MPa

Run no.	k (m/s)
A2	2.02E-05
A3	2.82E-05
A4	3.33E-05
avg	2.73E-05

Table D-2 mass transfer coefficient at 0.5 MPa

Run no.	k (m/s)
A2	2.01E-05
A3	2.18E-05
A4	1.05E-05
avg	1.75E-05

APPENDIX G

THE CONCENTRATION POLARIZATION: THE FLUORIDE CONCENTRATION ON MEMBRANE SURFACE (CM)

The fluoride concentration on membrane surface (C_M) was calculated from Equation (2-6).

$$\left(J_v\right)_{solute} = k \ln\left(\frac{C_M - C_P}{C_B - C_P}\right)$$

Where k = $\left(D/\delta\right)$= mass transfer coefficient, D = solute diffusion coefficient, δ = boundary layer thickness, $\left(J_v\right)_{solute}$ = permeate flux of solute solution, C_M = solute concentration prevailing on membrane surface, C_P = solute concentration in permeate solution, and C_B = solute concentration in bulk solution.

Example for run no. A2:

Run no.	Mass transfer coefficient (k), m/s	Concentrated water (C_B), mM	Permeate water (C_P), mM	Solute flux (m^3/m^2-s)
A2	$2.73\text{x}10^{-5}$	11.81	0.60	$6.061\text{x}10^{-6}$

$$6.061x10^{-6} = 2.73x10^{-5} \ln\left(\frac{C_M - 0.60}{11.81 - 0.60}\right)$$

C_M = 14.60 mM

APPENDIX H

GROUNDWATER DEFLUORIDATION EXPERIMENT: THE FLUORIDE CONCENTRATION ON MEMBRANE SURFACE (C_M) AND ON GEL LAYER SURFACE (C_G)

The fluoride concentration on gel layer surface was calculated from equation (2-6). By substitute C_M with C_G

$$\left(J_v\right)_{solute} = k\ln\left(\frac{C_M - C_P}{C_B - C_P}\right)$$

The fluoride concentration on membrane surface was calculated from equation (2-9)

$$J_i = k_i\left(C_M - C_P\right)$$

Where k_i = solute mass transfer coefficient, Ji = solute flux (Jv $_{(solute)}$. C_p), C_M = concentration of membrane surface, C_P = concentration of permeate

Example: Pra Too Khong groundwater at 250 ml permeate water obtained under 0.3 MPa

Permeate water obtained	Concentrated water (C_B), mM	Permeate water (C_P), mM	Permeate flux m3/m2-s	Mass transfer coefficient (k), m/s	Mass transfer coefficient through membrane (K), m/s
250 ml	13.99	0.55	$5.180\ x10^{-6}$	$2.73x10^{-5}$	$3.92x10^{-7}$

C_G calculation $$5.180x10^{-6} = 2.73x10^{-5}\ln\left(\frac{C_G - 0.55}{13.99 - 0.55}\right)$$

$$C_G = 16.80 \text{ mg/L}$$

C_M calculation $$2.85x10^{-6} = 3.92x10^{-7}\ (CM - 0.55)$$

$$C_M = 7.81 \text{ mg/L}$$

APPENDIX I

THE FLUORIDE REJECTION RATE

The fluoride rejection rate can be determined by following equation.

$$\%R = (1 - \frac{C_P}{C_B}) \times 100$$

Where %R= percent fluoride rejection rate, C_P = fluoride concentration in permeate water, C_B = fluoride concentration in concentrated water

Table I-1 Fluoride rejection rate of Pra Too Khong groundwater

Permeate water obtained (ml)	Transmembrane pressure (MPa)	Fluoride rejection rate (%)
250 ml	0.3	96.1
500 ml	0.3	95.9
750 ml	0.3	95.7
1000 ml	0.3	96.0
250 ml	0.5	96.7
500 ml	0.5	96.4
750 ml	0.5	96.1
1000 ml	0.5	96.3

Table I-2 Fluoride rejection rate of Pra Too Khong groundwater with 500 mg/L of silica concentration

Permeate water obtained (ml)	Transmembrane pressure (MPa)	Fluoride rejection rate (%)
250 ml	0.3	92.1
500 ml	0.3	92.3
750 ml	0.3	92.2
1000 ml	0.3	92.1
250 ml	0.5	94.6
500 ml	0.5	94.7
750 ml	0.5	94.8
1000 ml	0.5	94.1

Table I-3 Fluoride rejection rate of San Kam Pang hot spring well

Permeate water obtained (ml)	Transmembrane pressure (MPa)	Fluoride rejection rate (%)
250 ml	0.3	96.7
500 ml	0.3	97.1
750 ml	0.3	97.2
1000 ml	0.3	97.3
250 ml	0.5	97.4
500 ml	0.5	97.4
750 ml	0.5	97.5
1000 ml	0.5	97.5

Table I-4 Fluoride rejection rate of San Kam Pang hot spring well with 500 mg/l of silica concentration

Permeate water obtained (ml)	Transmembrane pressure (MPa)	Fluoride rejection rate (%)
250 ml	0.3	91.9
500 ml	0.3	92.1
750 ml	0.3	92.2
1000 ml	0.3	92.1
250 ml	0.5	93.3
500 ml	0.5	93.3
750 ml	0.5	93.1
1000 ml	0.5	93.3

BIOGRAPHY

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