Development and Evaluation of flux enhancement and cleaning strategies of woven fibre microfiltration membranes for raw water treatment in drinking water production

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Declaration

I hereby declare that this dissertation is my own work and to the best of my knowledge it neither contains materials previously published or written by another author and it has not been submitted in part or in whole for the award of another degree at Durban University of Technology (DUT) or any other educational institution. I also declare that the academic content of the dissertation is based on my work. All literature cited and contributions made by others have been acknowledged in this work.

Kumnandi Pikwa

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Signature

Abstract

Woven Fibre Microfiltration (WFMF) membranes have several advantages over its competitors with respect to durability, making it a favourable alternative for the developing world and operation during rough conditions. Wide application of membrane technology has been limited by membrane fouling. The durability of the WFMF membrane allows more options for flux enhancement and cleaning methods that can be used with the membranes even if they are vigorous. Therefore, the purpose of this work was to develop and evaluate flux enhancement and cleaning strategies for WFMF membranes.

Feed samples with high contents of organics and turbidity were required for the study. Based on this, two rivers which are Umkomaasi and Duzi River were identified to satisfy these criteria. A synthetic feed with similar fouling characteristics as the two river water was prepared and used for this study. The synthetic feed solution was made up of 2 g/ℓ of river clay in tap water and 0.5% domestic sewerage was added into the solution accounting for 2% of the total volume. A membrane filtration unit was used for this study. The unit consisted of a pack of five membrane modules which were fully immersed into a 100 litres filtration tank. The system was operated under gravity and the level in the filtration tank was kept constant by a level float. The study focused on evaluating the performance of the woven fibre membrane filtration unit with respect to its fouling propensity to different feed samples. It also evaluated and developed flux enhancement and cleaning strategies and flux restoration after fouling. The results were compared to a base case for flux enhancement and pure water fluxes for cleaning.

The WFMF membrane was found to be prone to both internal and external fouling when used in the treatment of raw water (synthetic feed). Internal fouling was found to occur quickly in the first few minutes of filtration and it was the major contributor for the loss of flux from the WFMF membrane. The fouling mechanism responsible for internal fouling was found to be largely pore blocking and pore narrowing due to particle adsorption on/in the membrane pores. The structure (pore size, material and surface layout) of the WFMF membrane was found to be the main cause that made it prone to internal fouling. The major fouling of the WFMF membrane was due to internal fouling, a high aeration rate of 30 ℓ/min had minimal effect on the fouling reduction. An aeration rate of 30 ℓ/min improved the average flux by only 36%, where a combination of intermittent backwashing with brushing and intermittent backwashing with aeration (aeration during backwashing only) improved average flux by 187% and 135% respectively. Pre-coating the WFMF membrane with lime reduced the effects of pore plugging and particle adsorption on the membrane and improved the average flux by 66%. The cleaning strategies that were most successful in pure water flux (PWF) recovery were high pressure cleaning and a combination of soaking and brushing the membrane in a 0.1% NaOCl (desired) solution. PWF recovery by these two methods was 97% and 95% respectively.

Based on these findings, it was concluded that the WFMF membrane is susceptible to pore plugging by colloidal material and adsorption/attachment by microbiological contaminants which took effect in the first hour of filtration. This led to a 50% loss in flux. Also, a single flux enhancement strategy proved insufficient to maintain a high flux successfully. Therefore, combined flux enhancement strategies yielded the best results.

Dedication

This dissertation is dedicated to my parents Samkelo Pikwa and Nodumo Pikwa for always believing in me and trusting me and never putting pressure on me. I also would like to dedicate this dissertation to my lovely wife Dorcas Alfa, Rev & Mrs J Abechi, M Chollom, Prof. V.L. Pillay and Dr. S. Rathilal who never gave up on me but continued to encourage me to complete the work even when I did not feel like.

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List of Abbreviations/Formulae

Calcium hydroxide – $Ca(OH)_2$

CP – Concentration Polarization

CFU – Coliform units

DUT – Durban University of Technology

EPS – Extracellular polymeric Substances

IMS – Integrate membrane systems

MF – Microfiltration

MSE – Modified size exclusion methods

NF- Nanofiltration

NOM – Natural Organic Matter

POU – Point Of Use systems

PWF – Pure Water Flux

RO – Reverse Osmosis

UF – Ultrafiltration

UNMDG - United Nation Millennium Development Goal

USEPA – United State Environmental Protection Agency

SA – South Africa

SMP – Soluble Microbial Products

TMP – Transmembrane pressure

WHO – World Health Organisation

WFMF – Woven Fibre Microfiltration

Chapter-1 INTRODUCTION

1.1 Water problems

Access to safe drinking water continues to be a challenge in developing countries. According to UNICEF (2008), one of the biggest killer of the 21st century particularly in children and old age, is waterborne diseases which results from unsafe drinking water and poor sanitation conditions (open defecation). The United Nation Millennium Development Goal (UNMDG) number 7C seeks to address this issue. Its revised targets are that 88.3% of the world's population should have access to improved sources of drinking water and that 74.6% of the world's population should have access to improved sanitation facilities by 2015. Data from the World Health Organisation shows that as of 2012 (no new data is currently available apart from the 2012 report by WHO on UNMDG). Only 53% in the sub-Saharan African countries had access to improved sources of drinking water and only 23% had access to improved sanitation facilities. This data was based on the rural communities only. These statistics spell out the disaster that awaits the poorest of the world communities living in the rural areas of sub-Saharan African countries.

Improving drinking water and sanitation facilities alone does not completely solve the problem of waterborne diseases. Treatment of drinking water is still essential and cannot be substituted with anything. According to Statistics-South Africa (2013) the UNMDG target for improved drinking water sources was achieved in 2005 in South Africa (SA) and the improved sanitation target lagged behind at 66%. Yet, in 2008, due to cross border and water contamination, an outbreak of cholera in SA was experienced, which claimed 57 lives and about 12000 infections were recorded (Blumberg et al. 2011). Therefore, improving drinking water sources and also sanitation is critical.

1.2 Conventional water treatment

Conventional water treatment facilities are a combination of different treatment processes commonly arranged in a train sequence. A typical process for conventional water treatment plants includes: screening (course-filtration), flocculation, sedimentation, filtration and disinfection. Conventional water treatment facilities are capable of producing quality drinking water when operated within the confines of their design. However there are fundamental challenges that arise if conventional water treatment is to be used as the vehicle to meet the UNMDG goal number 7C which states that by 2015, the proportion of people without sustainable access to safe drinking water and basic sanitation should be halved. These challenges include;

- ✓ Huge capital costs in the hundreds of millions of Rands for new equipment or refurbishment of the currently aged equipment. The improvement of drinking water sources in SA has come with a lot of debt from the World Bank (Statistics-South-Africa, 2013).

- ✓ The cost of erecting dams is high since these systems require large volumes of raw water for storage proper functionality.

- ✓ There is a high level of pollution from water sources which current water treatment facilities were not designed for. Some of these contaminants are due to industrialisation, urbanisation and disposals from agricultural pollutants especially in developing countries. According to Appleton and Bailey (1990) wastewater works are not coping with the increase in the pollution load and therefore, there is usually an overflow to rivers.

- ✓ Difficult topography in the rural areas and lack of development has limited the installation and distribution of clean water.

All these challenges have made it almost impossible to supply piped drinking water to people in the rural communities. South Africa managed to improve drinking water sources to about 89% of its population 10 years ago (Statistics-South-Africa, 2013). Since then there has not been any improvement in the number of people receiving piped drinking water because of those challenges. Again, one of the major factors that limit and increase the cost of conventional water treatment facilities is their centralized nature (Peter-Varbanets *et al.* 2009). Decentralized and stand-alone systems such as point of use units (POU) could be a viable option to achieve the UNMDG goal 7C. The main purpose of treating drinking water is to bring it to an acceptable level of quality for human consumption, by removing microorganisms (bacteria) and viruses that could cause harm to human health. The removal of these dangerous microbes in the conventional water treatment process is achieved mostly in the filtration and disinfection processes (USEPA, 2004).

With the use of alternative advanced methods of water treatment, the gaps in the conventional (centralised) water treatment facilities can be covered. One such alternative is the use of membrane technology. This is evident by the tremendous amount of growth this technology has enjoyed both in research work and application in the recent years. Madaeni (1999) explained that membranes can increase the safety of water in two ways: firstly, they can be used at the consumption point as a tool for increasing security and secondly they can be part of the water purification system.

1.3 Membrane technology

Membrane technology is a generic name used when referring to the separation process that uses a membrane. The use of membrane filtration process for the treatment of potable water at household levels has become more attractive recently due to increasing stringent rules in drinking water guidelines. The superiorities of membrane technology over the conventional water treatment plants include but is not limited to; flexibility to upgrades and scaling, ability to handle varying incoming quality, small foot print plants, can be decentralized and centralized at will, can be easily incorporated into existing water treatment facilities and produce superior quality than most of the competing technologies (Peter-Varbanets *et al.* 2009). The advantages offered by membrane technology has made it suitable for rural communities where topographies are difficult and there is no infrastructure that conventional water treatment plants require (Lee *et al*. 2011).

1.4 Membrane technology limitations

Wide application and research of membrane technology has vastly been carried out towards domestic wastewater treatment and industrial wastewater treatment. Streams from these sources are well researched and understood. They do not vary greatly from one plant/area to the other, thereby making wastewater treatment facilities less troublesome to design, operate and even research. However, the raw water for drinking water treatment facilities differs from one area to the other and from one season to the other. Therefore, the plant design requirements for specific waters are different to others, even research findings are specific to the water studied (Nicolaisen, 2002; Carroll, 2009). Lack of accurate information on the quality of the raw water to be used with the membrane at design stages has enhanced the risk of membrane fouling in drinking water treatment. Membrane fouling is a major concern in drinking water treatment systems because high costs and membrane integrity issues are incurred whilst trying to maintain the membrane. Commercially available membranes are very sensitive to rough treatment resulting in frequent cycles and maintenance costs (Pikwa *et al.,* 2010). Membrane fouling contributes to 80% of the operational and maintenance costs of membrane systems; this is even higher than the skills of the personnel operating membrane systems. Operation and maintenance costs of membrane systems could be higher than the capital costs of the system.

1.5 Fouling

Fouling is inherent to all filtration systems including membranes. Membrane fouling is the deposition of solutes being filtered by the membrane on the membrane surface and its internal pores. The deposited solutes can chemically attach to the membrane and cause structural change of the membrane. This often results in a reduction in flow through the membrane, and when this happens the membrane is fouled (Goosen *et al.*, 2004; Hwang *et al.,* 2008). The longer the membrane is allowed to operate in its fouled state, the harder it becomes to remove the foulants from the membrane. It therefore becomes necessary to use chemicals or greater force to recover a highly fouled membrane. This results in membrane damage and or shortened life span of the membrane. This is because most of the available membranes are sensitive to the cleaning chemicals. One of the root cause of the problems experienced with cleaning the membranes with chemicals, lies in the fact that the cleaning procedures and chemicals used are provided by the membrane manufacturers. The procedures for cleaning used by the manufacturers are usually based on a different feed, with different fouling characteristics to that which the user usually applies. So when a cleaning methodology is provided by the membrane supplier it does not give satisfactory results with those of the users. Therefore, a deviation in the expected outcomes is experienced and as a result, the membrane is sometimes damaged.

Current available membranes are not suitable for rough treatments and as a result the user needs to keep strictly to the maintenance procedures supplied by the manufacturers if the full benefits of the technology are to be enjoyed. This was identified as a gap in the technology by the Department of Chemical Engineering, Membrane Water Research Group at Durban University of Technology. This then led to the evaluation of a woven fibre micro filter membrane (WFMF). The membrane is made of a polyester based fibre material, locally supplied by the Gelvenor Consolidated Fabrics (PTY) Ltd in Durban, South Africa. The membrane has the following advantages to offer:

- ✓ Has a high mechanical and tensile strength
- ✓ Is robust
- ✓ Is inexpensive
- ✓ Readily available locally
- ✓ This membrane is suitable for under developed countries where the technical know how to operate and handle complex and sensitive systems is limited. The separation integrity of the material was found to be satisfactory for the purpose of drinking water treatment (Pikwa *et al.,* 2010).

1.6 Focus of the study

This project focused on evaluating the performance of the woven fibre membrane filtration unit with respect to its fouling propensity to different feed samples. It also evaluated and developed flux enhancement and cleaning strategies and flux restoration after fouling.

1.7 Feed selection and approach

Feed samples with a high content of organics and turbidity were required for the study.The two rivers which are the Umkomaasi and Duzi River were identified to satisfy these criteria. These rivers are located 60 and 90 kilometres respectively from Durban University Technology (DUT) where the study was conducted. It was not feasible to set up the study close to the river and very tedious as well for a continuous supply of river water to DUT, a synthetic feed with similar fouling characteristics as the two rivers water was prepared and used for this study. The synthetic feed solution was made up of 2 g/ℓ of river clay in tap water and 0.5% domestic sewerage was added into the solution accounting for 2% of the total volume. The turbidity of the synthetic feed was 200 ± 5 NTU and the *E.coli* count was 12000 ± 100 CFU. The fouling profile of the synthetic feed was similar to that of Duzi River which at the time of the test had a turbidity of 150 NTU and *E.coli* count of 8412 CFU.

The system was operated under gravity and the level in the filtration tank was kept constant by a level float. The experiments for this study were divided into three.

(1) WFMF membrane and feed solution fouling characterization

The experiment was to ensure the membrane was given a constant continuous separation integrity by evaluating the turbidity of the membrane before filtration commenced and regularly thereafter. A good WFMF membrane produces a filtrate turbidity of less than 1 NTU for the solution that is filtered. The most important turbidity check was done immediately after membrane cleaning. This was to verify that no damage was made to the membrane during cleaning. Fouling characterization experiments were conducted to determine the flux decline rate under constant flux operation. Six different samples were filtered and the performance of the WFMF membrane with respect to flux decline was observed. The samples used were Duzi River, Umkomaasi River, Synthetic feed, clay solution, 0.8 µm calcium hydroxide ($Ca(OH)_{2)}$ and 2 µm Ca(OH)2 solution.

(2) Assessment of various flux enhancement strategies on the WFMF membrane

The performance of each flux enhancement strategy was measured against the base case. For each flux enhancement experiment the base case fouling profile was plotted. The base case profile indicates the lowest flux possible for the specific feed, WFMF membrane and operating conditions. Two external membranes in the membrane pack were used to plot the base case curve.

(3) Optimization of the flux enhancement strategies and assessment of cleaning strategies for the WFMF membrane.

Cleaning was determined by the pure water flux (PWF) recovery after cleaning. The PWF of a cleaned membrane was compared to that of a new membrane as it is expected that that the flux decline profile of a cleaned membrane was expected to be similar to that of a new or clean membrane.

1.8 Objectives of the study

The objectives of the study are divided into: (1) Evaluating and developing the WFMF membrane and feed solution fouling characterization (2) Assessment of various flux enhancement strategies on WFMF membrane (3) Optimization of the flux enhancement strategies and (4) Assessment of cleaning strategies for the WFMF membrane. The detailed breakdown of these is listed in the subsequent sections.

Evaluating and developing WFMF membrane and feed solution fouling characterization.

- ✓ Evaluating the fouling rate with respect to time
- ✓ Evaluating irreversible fouling in the WFMF
- ✓ Developing a gradient to assess fouling rate and flux recovery after cleaning

Investigate flux enhancement of the WFMF membrane

- ✓ Aeration

- ✓ Backwashing
- ✓ In-situ brushing
- ✓ Flux control (operate the system below critical flux)
- ✓ WFMF membrane pre-coating

Develop and evaluate cleaning strategies for the WFMF membrane

- ✓ Chemical cleaning and rinse with tap water
- ✓ Flush with tap water after filtration
- ✓ External brushing with tap water
- ✓ Chemical soaking and brushing
- ✓ Cleaning with commercial soap and brushing
- ✓ Pressure cleaning

Chapter-2 LITERATURE REVIEW

2.1 Water pollution

Water is generally referred to as a universal solvent, implying that it has the ability to dissolve anything it is in contact with, if given sufficient time. It is this solvent nature that makes it unsafe if necessary steps of analysis and treatment are not undertaken before it is used for human consumption and related uses.

Raw water sources for water treatment facilities can either be surface water or ground water. Surface water sources include rivers, lakes and reservoirs, while ground water is from boreholes, springs and wells (Sobsey, 2002). Contaminants in raw water are dependent on the source and the activities around the source. Ground water is usually expected to be safe for consumption requiring no further treatment. However, external human activities around ground water sources could contaminate the water, rendering it unsafe for drinking if not treated (Brikké and Bredero, 2003). Surface water is exposed to all kinds of activities such as, wet weather run-off, atmospheric and human activities. These activities make surface water susceptible to contamination and must be treated before consumption according to Safe Drinking Water acts (WHO, 2011 SANS 241, 2011).

Raw water is so infinitely variable in quality that it is difficult to standardize the treatment process to a fixed starting point and finishing point. Therefore, starting and finishing points of any water treatment facility is determined by the contaminants found in such water (Smethurst, 1979). Not all contaminants found in raw water are dangerous for human use. To attempt to measure and remove every contaminant in raw and treated water is unjustified and costly. Typical parameters/contaminants that are measured and treated to acceptable limits according to Cheremisinoff (2002) and Binnie and Kimber (2009) are listed in Table 2-1. These parameters vary from one source to another and from one water treatment facility to the other. Table 2-1 shows the most typical parameters.

Table 2-1 Typical raw drinking water contaiminants (Binnie and Kimber, 2009; Cheremisinoff, 2002)

Parameter	**Units**
Colour	Hazen
Turbidity	NTU
pH	
Conductivity	µm/s
Hardness	mg/l
Total Nitrogen	mg/l
Chemical Oxygen Demand	mg/l
Biological Oxygen Demand	mg/l
Total Coliforms	CFU
Faecal Coliforms	CFU

Contaminants in raw water exist in three progressive fine states i.e., suspended, colloidal and dissolved matter. The primary objective of a water treatment facility is the removal of microbiological parameters (Coliforms) which pose a major threat to human health (Binnie and Kimber, 2009). Incorporated under turbidity is a wide range of parameters such as suspended solids, colloidal matter, heavy metals, organic matter and microorganisms. Therefore, turbidity is also a very important parameter to measure because it can be used to detect the presence of other harmful substances in water other than microorganisms. According to Cheremisinoff (2002), turbidity is bad for aesthetic considerations and that turbidity decreases the effectiveness of water treatment techniques by shielding pathogens from chemical or thermal damage, shielding pathogens from chemical attack and absorbing the (ultraviolet) UV light, in case of *UV* treatment.

2.2 Water treatment

Potable water treatment can primarily be divided into; house-hold treatment and conventional water treatment. House-hold water treatment in nature has, or should have, minimal technical requirements and expertise. Regulation of the house-hold systems is almost non-existent due to the lack of monitoring, assessment and standardization (Brikké and Bredero, 2003). House-hold water treatment systems include the boiling of water, slow sand filtration and domestic chlorination. Boiling of water according to WHO (2005) kills or inactivates microorganisms but it negatively affects the taste of the water and does not improve the aesthetic state of the water. Slow sand filtration involves passing raw water through a bed of sand where it is treated by both biological and physical treatment.

The fine particles that cause turbidity are trapped by the bed of sand. Included in the trapped particles are microorganisms. Growth of substances such as microorganisms on top of the filter bed is crucial in slow sand filtration, because they offer biological treatment of the raw water as it passes through the sand filter by feeding on the bacteria coming in with the raw water, thereby producing bacteria-free water. House-hold chlorination involves addition of a pre-determined amount of chlorine to a measured amount of water being treated. This process kills bacteria present in water; however, chlorine addition introduces taste and odour in the water and this hinders the continuous use of chlorine (Brikké and Bredero, 2003).

2.2.1 Conventional Water Treatment

Conventional water treatment facilities are a combination of different treatment processes commonly arranged in a train sequence. A typical process for conventional water plants includes; screening (course-filtration), flocculation, sedimentation, filtration and disinfection. Depending on contaminants found in specific water, other processes such as ion exchange, adsorption, reverse osmosis, etc. can be included in the treatment train (USEPA, 2004). Table 2-2 shows the typical processes for conventional water treatment.

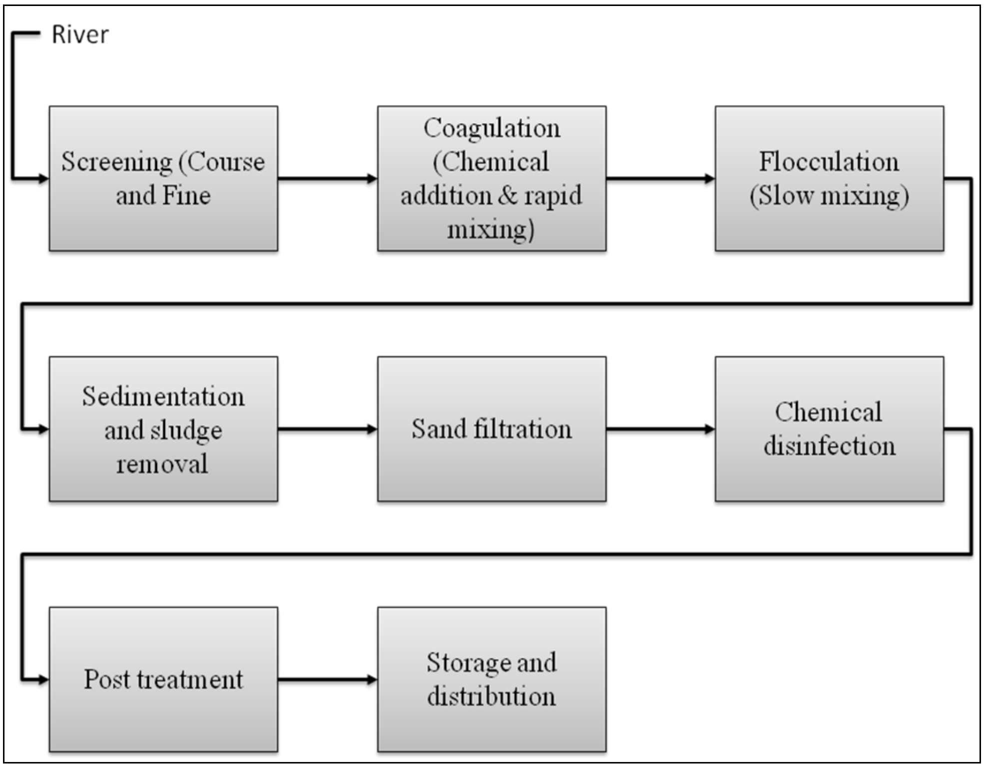

Figure 2-1 Conventional water treatment process (Cheremisinoff, 2002)

The conventional water treatment process according to USEPA (2004) begins with water screening from the source. The screening process deals with the removal of large particles present in water like rags, logs, plastics, etc. Screening is however more prevalent in wastewatertreatment, but is not foreign in drinking water treatment and the need for screening is largely dependent on the source of the water and how protected the source is. Screening out large particles is for protection of equipment from wear and tear and preventing equipment blockages resulting from the presence of these particles.

Screening is followed by coagulation, where the coagulation chemical such as ferric sulphate and polyelectrolyte are added at a predetermined dosage. Care must be taken that the pH is correct for coagulation to take effect. Directly after the addition of the coagulants, vigorous mixing takes place to ensure that efficient and complete mixing of coagulants and water occurs. In coagulation, coagulants neutralize the electrical charges of particles in water that prevent coagulation. Once the charges are neutralized, the fine particles begin to clump together (Bratby, 2006). A study by Ayguna and Yilmazb (2010) revealed that depending on the contamination of the water, a single combination of coagulants and coagulant aids may not be adequate. Thus a combination of coagulants could be necessary in such cases. This is true for water that is heavily contaminated with domestic waste. Coagulation plays a major part in conventional water treatment in the sense that it affects two solid removal processes. These are sedimentation and filtration. It can also affect the chemical disinfection process. Inefficient coagulation has a negative impact on filtration and disinfection. Poor coagulation could lead to frequent plugging of the filter bed, while also leading to inefficient removal of turbidity in the filtration process and the turbidity passing to the disinfection step. Complete disinfection can be hindered in the presence of turbidity (Kerri, 2008). A study by Giani *et al.* (2011) showed that efficient coagulation is dependent on the incoming turbidity, pH and coagulant dose. Increasing these three parameters improves the efficiency of coagulation, with respect to final turbidity removal.

Coagulation is followed by flocculation, which is typically done in large basins with gentle mixing. Flocculation is a process where the fine particles produced during coagulation clump together into bigger and denser flocs that are easy to separate by settling. Rapid mixing, coagulation and flocculation can be combined in a large single basin with compartments for each process. The main objective of these three processes is to remove turbidity which will otherwise have a negative impact on disinfection. Also removed by these processes is colour and smell. The factors that are monitored during coagulation and flocculation are adequate rapid mixing, sufficient doses of coagulants, proper gentle mixing, correct pH and temperature (Bratby, 2006). Flocculation as studied by Gidde and Bhalerao (2010) showed that there is an optimum slow mixing velocity of 60 revolutions per minute (rpm), above which re-dispersion of particles that had already formed flocs may occur. Also shown in their study was that low incoming turbidity requires low mixing velocity, while high incoming turbidity requires high mixing velocity.

Sedimentation follows flocculation but there are cases where pre-sedimentation is done prior to coagulation and flocculation. This is typically done to reduce the load on the coagulation and flocculation, hence a reduction in the demand of coagulating chemicals. During sedimentation the velocity of water is reduced to be lower than the suspension velocity thereby allowing suspended solids to settle out due to gravity. Sedimentation is used to separate out the large flocs formed during the previous processes. Typically about 90% of the suspended particles and bacteria are removed during sedimentation thereby reducing the load for the filtration process. The sedimentation basin has four main zones or parts; entry zone, settling zone, exit zone and de-sludging zone.

If proper control and operation is not taken in each zone, sedimentation efficiency is negatively affected. In the entry zone, water is distributed evenly at a slow velocity to ensure that there is no short-circuiting. In the settling zone, most of the separation takes place. The liquid velocity in this zone must be brought down to less than 0.5 m/s, otherwise the flocs break and get suspended in water reducing the sedimentation efficiency. Like the entry zone the exit zone is also used to prevent short circuiting and prevents flocs from exiting with the water.

From the de-sludging zone all the flocs that have settled to the bottom of the basin are removed, otherwise these will break and be suspended back into the water and can become septic (Sobsey *et al.,* 2008). Sedimentation according to Hasselblad *et al.* (1998) occurs in four stages, discrete particle settling, flocculent settling, hindered settling and compression. The discrete settling is governed by Newton and Stokes laws. Flocculent settling can be explained by the neutralization effect of charges offered by coagulants after which particles flocculate and form bigger particles that settle according to discrete settling. Hindered settling as explained above is based on the solid flux theory. This settling is determined by the gravitational velocity and bulk concentration, meaning a faster rate of de-sludging results in better zone settling. The Stokes law corrected for laminar flow during sedimentation is shown in equation 2-1

$$v = \frac{g(\rho_p - \rho_f)d^2}{18\mu} \qquad \textbf{2- 1}$$

Where v is the flow velocity (m/s), p_p is the particle density (kg/m^3), p_f is the fluid density (kg/m^3), d is the particle diameter (m) and u is the dynamic viscosity (kg/m.s)

Newton second law is;

$$m\frac{dv}{dt} = F_g - F_t - F_f \qquad \textbf{2- 2}$$

The gravity force F_g is given by:

$$F_g = mg = \rho_p V_p g \qquad \textbf{2- 3}$$

The lifting force F_t is given by:

$$F_t = \rho_f V_p g \qquad \textbf{2- 4}$$

The frictional drag force F_f is given by the following equation and is a function of particle velocity, fluid density, projected area and drag coefficient;

$$F_f = \frac{C_D A_p \rho_f v^2}{2} \qquad \textbf{2- 5}$$

C_D is a dimensionless drag coefficient; A_p is projected area (m^2); ρ_f is fluid density (Kg/m^3); v is particle velocity (m/s); V_p is the volume (m^3); g is the gravitational force (m/s^2); m is the mass (kg); ρ_p is the particle density (kg/m^3).

Filtration is the final step of solid removal in water treatment, and it is mandatory for all water treatment processes (Hespanhol and Prost, 1994). This is so because there are bacteria that cannot be disinfected by chemicals but need physical removal. Another major purpose of the filtration step in water treatment is turbidity control which should be less than 0.5 NTU when leaving the filters and should never be above 5 NTU at the disinfection stage (USEPA, 2001). Conventional filtration involves passing of water which has advanced from coagulation, flocculation and sedimentation through a bed of sand. Sand bed filtration/granular separation occur within the granular medium (depth filtration) and rarely on the top layer (cake filtration/strain filtration). Filtration occurs also by the adsorption mechanism. This is influenced by the type of filtration bed used, i.e. presence of coal in the bed will cause adsorption.

The size of flocs in the filter affects the operation of the filter and its efficiency. In the event that small flocs are sent to the filter bed, its removal is poor, resulting in high turbid effluent from the filters, while big flocs and high solid load to the filter bed, results in a frequent plugging of the filter. Continuous monitoring of the filtrate quality and pressure loss across the filter is necessary for good operation of sand filters (Bratby, 2006). A review of conventional filtration by USEPA (2001) showed that proper operation of granular filters used in conventional water treatment can result in 4 log removal of protozoan pathogens. The review showed that granular filtration is affected by the size and density of the microbes, size and depth of the filter medium and filtration rate. Efficient granular filtration needs to be preceded by proper pre-treatment. The maintenance of the filter is vital and it involves mainly backwashing of the filter periodically at predetermined time intervals or when there is a significant pressure drop across the filter or breakthrough of turbidity.

The critical part of disinfection is the removal or inactivation of disease causing organisms that are present in raw water. As early as sedimentation and filtration disinfection already takes place when substances such as *Giardia* are removed through settling and size exclusion in the filtration process. *Giardia* is amongst the protozoa that cannot be chemically inactivated but requires physical removal (USEPA, 2000). The disinfection step which, in conventional water treatment is usually chlorination can be replaced by either nanofiltration or reverse osmosis. However, some water bodies such DWAF, USEPA, etc. mandates that chlorination must be done as post treatment for residual purpose to prevent recontamination. Chlorine related chemicals are added to the water to kill/inactivate the remaining bacteria in the water after sedimentation and filtration. The presence of turbidity in water at the disinfection step affects the chlorine demand required to disinfect water (Schoenen, 2002). According to Kerri (2008) and many other disinfection researchers such as Schoenen (2002), Cheremisinoff (2002) and Momba and Brouckaert (2005) found that the presence of turbidity in water at the disinfection step hinders the efficiency of disinfection. This is the reason great care is taken in the upstream processes to the disinfection process.

According to Bilqis *et al.* (2006), in most conventional water treatment processes, chemical disinfection is the last step before consumption, so care must be taken to make sure it works. Ineffective pre-treatment upstream affects disinfection kinetics i.e. dose, residence time, agitation (Schoenen, 2002).

2.2.2 Limitations of conventional water treatment

Conventional water treatment plants consist of a long train of processes working properly in order to maintain reasonable quality drinking water exiting the plant. The maintenance and/or upgrading of existing processes with the same unit is costly and unnecessary (Fan *et al.,* 2014)**.**

The size of conventional water treatment plants require large storage feeders and are very centralized processes. The cost that is inherent to centralized systems is the redistribution cost, which in the case of a water treatment facility is in the form of pumping and piping costs. Also, the operating costs of conventional water treatment plants are quiet high (Varbanetsa *et al.,* 2008).

Conventional water treatment plants needs more instrumentation are not flexible to handle varying incoming water quality because these plants are designed and constructed for a specific feed quality with very little versatility. With industrialization and urbanisation, conventional water treatment feeders get highly contaminated by the increased human activities around the feeders and with new contaminants. The rigidity of conventional water treatment plants make them incapable to deal with the varying, high levels of contamination. Another limitation of conventional water treatment plants is the difficulty in adding new processes that could easily handle new contamination levels (Wei Lun *et al.,* 2014).

2.3 Alternative technology to conventional water treatment

The main purpose of treating drinking water is to bring it to an acceptable quality level for human consumption, by removing microorganisms (bacteria) and viruses that could cause harm to human health. The removal of these dangerous microbes in conventional water treatment processes is achieved mostly in the filtration and disinfection processes

(USEPA, 2004). According to Kerri, (2008) and the Environmental Protection Services Agency (EPA), the presence of turbidity during chemical disinfection reduces the effectiveness of disinfection. Organisms such as *Cryptosporidium* and *Giardia* are mandatory that they be physically removed by some form of filtration. *Cryptosporidium* and *Giardia* are common in surface water and are very dangerous to human health, therefore needs to be completely removed from drinking water (USEPA, 2001). By conventional filtration, 2 log removal of *Cryptosporidium* is achieved provided coagulation prior to filtration is optimized. Membrane filtration is able to achieve up to 6 log removal of *Cryptosporidium* which is equivalent to total removal, and this is without coagulation. Membrane technology can, therefore, be a viable alternative to conventional water treatment.

2.3.1 Membrane technology

Membrane technology is a physical separation process, where a membrane serves as a filter to separate a mixture. There is no modification of the substances being separated. The separation is through a membrane using momentum such as a pressure difference, concentration difference, or potential difference (Pinnekamp, 2003). There is a wide application of this technology i.e. manufacturing, medical, water treatment and fuel cells. A membrane according to Ho and Sirkar (1992) is an interphase between two bulk phases. The interphase can be one or a combination of non-porous solid, micro-porous or macro-porous with fluid in the pores. The purpose of the interphase (or a membrane) in between the two bulk phases is to control the exchange of mass transfer between the two bulk phases. The bulk phase is a mixture of species; during the membrane filtration process one or more species in the mixture is allowed to be exchanged in preference to the others. During this process one bulk phase is enriched in one or more of the species while the other is depleted of that species.

A membrane is defined by what it does and not what it is, hence a variety of materials can be used as membranes and because separation mechanism of a membrane does not define a membrane, several choices of membranes exist (Chang *et al.*, 2002). Figure 2-2 shows a membrane filtration process.

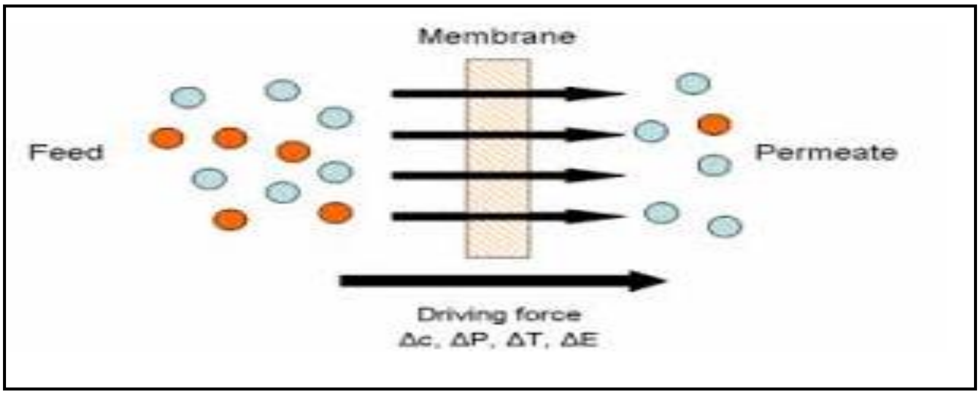

Figure 2-2 Membrane filtration process (adopted Membrane operations, 2014)

2.3.2 Membrane filtration process

A membrane filtration process is typically defined as a pressure or vacuum driven separation process in which particulate matter larger than 1 μm is rejected by an engineered barrier. The rejection is primarily through a size exclusion mechanism. The efficiency of the removal of targeted organism must be able to be quantified and verified through the application of a direct integrity test (Allgeier, 2005). The need to define membrane filtration came after the acceptance of MF and UF in water treatment. This was necessary in order to guide the technology developers and protect the users of the water produced from membrane systems (USEPA, 2001).

In a membrane filtration process for drinking water treatment, the feed water/raw water is pressurized towards the membrane cell, flowing either perpendicularly or parallel to the membrane. The high pressure in the feed water forces the water to pass through the membrane and the solutes are retained. The retained solutes form a concentration gradient on the membrane surface. This phenomenon is called concentration polarization and it leads to membrane fouling which affects the separation process. Membrane filtration systems are divided into two; low pressure filtration systems and high pressure filtration systems. The operating conditions of these processes are different and they include; driving pressure, concentration polarization and fouling layer development and feed quality constraints (Radcliff and Zarnadze, 2004). Figure 2-3 shows membrane filtration processes and the separation abilities.

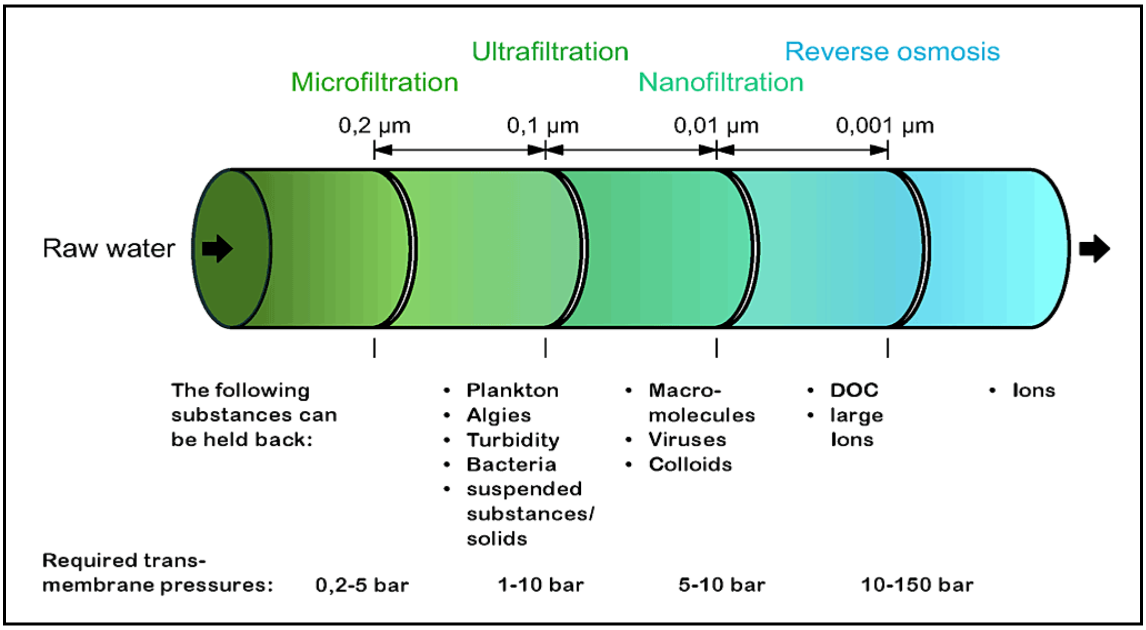

Figure 2-3 Membrane filtration processes and separation abilities (adopted from Hydro group-manual, 2012)

2.3.3 Low pressure membrane processes

MF and UF fall under the low pressure membrane processes. Their operating pressures range from 0.2 to 10 bars for MF and UF respectively. The pore sizes of MF starts from 0.1 to 0.2 µm. MF and UF are characterized by their ability to remove suspended and colloidal particles using a sieving mechanism based on size exclusion. UF pore sizes range from 0.1 to 0.01 µm. Some UF membranes can remove large organic macromolecules; these are the UF membranes whose separation ability has been characterized by molecular weight cut-off (MWC) and not pore size. Such organic macromolecules are morphologically difficult to define and are in a solution and not dispersed; hence, it becomes convenient in conceptual terms to use molecular weight cutoff (MWCO) rather than pore sizes (Allgeier, 2005).

MF can however use either depth filtration or screen filtration mechanisms. In depth filtration mechanism, the membrane pores on the surface are larger than the particles being filtered. So the particles are allowed to enter into the interior of the pore/channel. Whilst travelling the particle(s) get captured at the constrictions of the pore channel or get adsorbed in the wall of the pore channel. In screen filtration mechanism, the membrane pores are smaller than the particles being filtered, they then get trapped on the surface of the membrane due to size exclusion. Depth filters are generally thicker than screen filters; the reason being that with depth filters the thickness of the filter plays a separation role in the filtration process.

On the other hand, UF uses only screen filtration where suspended particles in a solution are rejected on the surface of the membrane due to size exclusion. The problem experienced in UF is concentration polarization while MF suffers mostly from pore clogging. Both of these problems lead to loss of production in the membrane (Baker, 2004). The filtrate quality from MF and UF is consistent and high when compared to conventional filtration systems (Karlapudi *et al.*, 2012). This is the reason MF and UF are now increasingly being used as pretreatment processes for NF and RO in both desalination and wastewatertreatment (Pilutti *et al.*, 2003).

2.3.4 High pressure membrane processes

High pressure membrane processes include NF and RO. The operating pressures for these processes range from 5 to 20 bars for NF and 10 to 100 bars for RO respectively. They are used mostly to remove dissolved contaminates. NF processes fall in the transition between pure RO process that operates at high pressures and UF process that operate at low pressures. NF can be called loose RO or low pressure RO (Baker, 2004).

Both NF and RO processes are designed to remove dissolved matter through a process of reverses osmosis. Separation by osmosis is driven by concentration differences, where the solvent moves through a semi-permeable membrane/barrier from high concentration to low concentration till an equilibrium state is reached. The pressure that drives the osmosis process is called osmotic pressure (Radcliff and Zarnadze 2004; Allgeier, 2005).

In reverse osmosis, the natural osmosis process is reversed by exerting on the feed mixture a pressure in excess of the osmotic pressure. The pressure on the feed side forces the solvent to pass through the semi-permeable membrane to solvent rich side of the membrane, thereby increasing the concentration of the solids on the other side of the membrane (i.e. feed side). The operating pressure of both NF and RO are affected by the amount or concentration of dissolved solids in the feed (Allgeier 2005). When NF and RO are used together in tandem, the function of NF is to soften the water whilst the RO's function is to polish the removal of dissolved matter and hardness (Samhaber *et al.*, 2009). Figure 2-4 shows a schematic representation of an osmosis and reverse osmosis processes, Table 2-2 compares the different membrane processes.

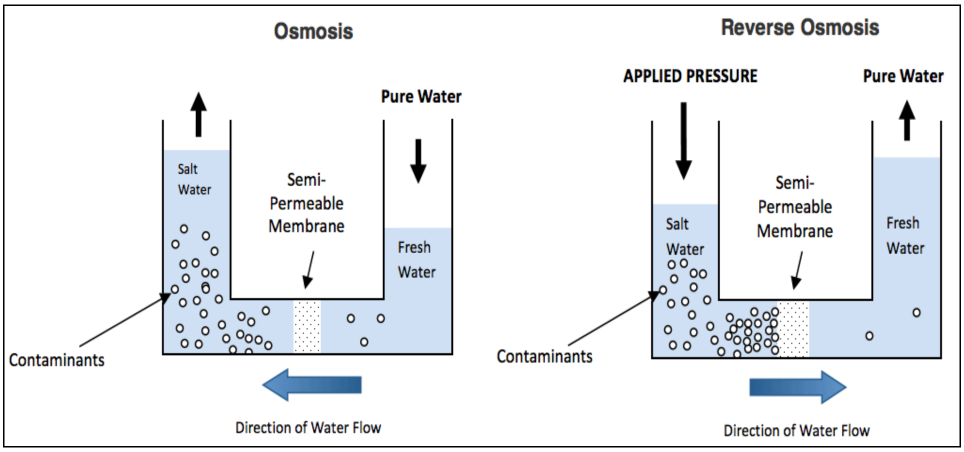

Figure 2-4 Schematic display for osmosis and reverse osmosis processes (EPA, 2005)

Table 2-2 Comparison of different membrane processes (adopted from Ravanchi *et al.,* 2009)

Membrane process	**Driving Pressure (bar)**	**Separation Principle**	**Entities Removed**	**Application**
Microfiltration	0.1 – 2	Sieving mechanism	Particles, suspended solid, larger bacteria	Water treatment, sterilisation, cell harvesting
Ultrafiltration	1 – 5	Sieving mechanism	Macromolecules, bacterial, fine colloidal material	Water treatment, textile, dairy & food, metallurgy
Nanofiltration	5 – 20	Solution diffusion	Low molecular salts, glucose, lactose, micro-pollutants	Desalination of brackish water, water softening, wastewatertreatment
Reverse Osmosis	10 – 100	Solution diffusion	Low molecular salts, glucose, lactose, micro-pollutants	Desalination of brackish and sea water, ultra-pure water production, wastewatertreatment

2.3.5 Stand-alone membrane system

The fact that a single membrane process is able to replace the entire conventional treatment train has made membrane technology very attractive in water treatment. According to Wei Lun *et al.* (2014), stand-alone membrane systems can either be a single membrane process i.e. microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) or even reverse osmosis (RO) or a combination of any of these processes. A single membrane process is more suitable for fairly clean raw feed with minimal fouling propensity. Fouling is a major problem in membrane systems and has limited the wide application of this technology (Van Agtmaal *et al.,* 2007). There are cases where a single membrane system is inadequate to address all the drinking water treatment requirements. For example microfiltration and ultrafiltration are poor in the removal of dissolved particles and natural organic matter (NOM) from water (Allgeier, 2005). These pollutants are easily removed by NF and or RO. However, if NF or RO is used alone without pre-treatment to remove colloidal matter and microorganisms that are deposited and forms biofilms on the NF or RO membranes, the net result is excessive fouling of these membranes, and the operation becomes excessively expensive (Van Paassen *et al.,* 1998; Van Agtmaal *et al.,* 2007; Di Profio *et al.*, 2011; Wei Lun *et al.,* 2014).

2.3.6 Integrated membrane systems

Integrated membrane systems (IMS) are processes where one or more membrane processes or non-membrane processes, are coupled with a membrane process into a single system. The purpose is to yield better performance of the integrated system, than from any of the individual components. The weakness of a one process is reduced by the strength of the other in an integrated system (Wei Lun *et al.,* 2014).

In most IMS the final process is usually RO. This process is normally used for final quality control of the product water. RO is able to bring drinking water to acceptable quality for desired usage. The requirement to integrate RO with other processes up stream is mainly for fouling control/mitigation. Fouling of RO membranes manifest itself as production loss due to scaling, organic fouling, particle deposition and biofouling (Van Paassen *et al.,* 1998). The fouling components that quickly foul NF/RO are easily removed by upstream processes without such high level of fouling to the upstream processes (MF and UF). A typical example of an IMS is water treatment in the Netherlands Heemsterk Water Treatment Plant where chemical disinfection/chlorination is replaced with reverse osmosis and the clarification process (coagulation, flocculation, sedimentation and filtration) is replaced by a single ultrafiltration process. Options to integrate NF/RO processes include conventional clarification, microfiltration, ultrafiltration and microfiltration/ultrafiltration with coagulation. Figures 2-5 to 2-7 show where the IMS system can be integrated for the treatment of water.

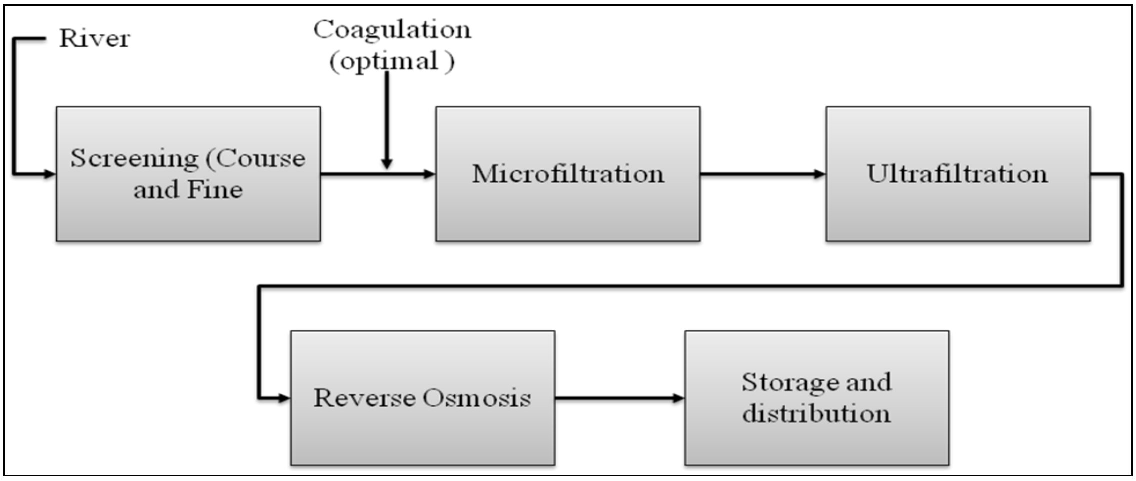

Figure 2-5 Integrated membrane system for high fouling water (adopted from Bick *et al.*, 2012)

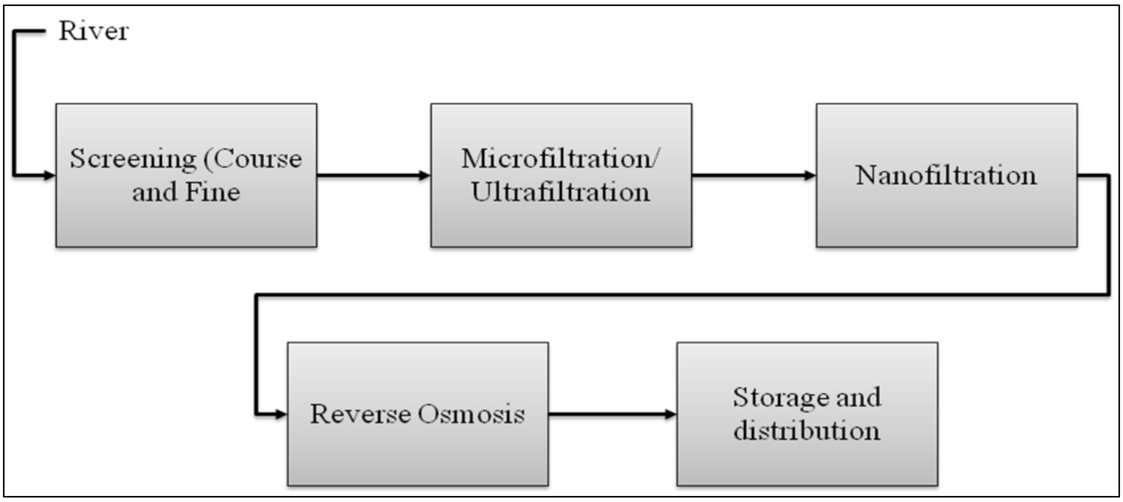

Figure 2-6 Integrated membrane system for hard water (adopted from Bick *et al.*, 2012)

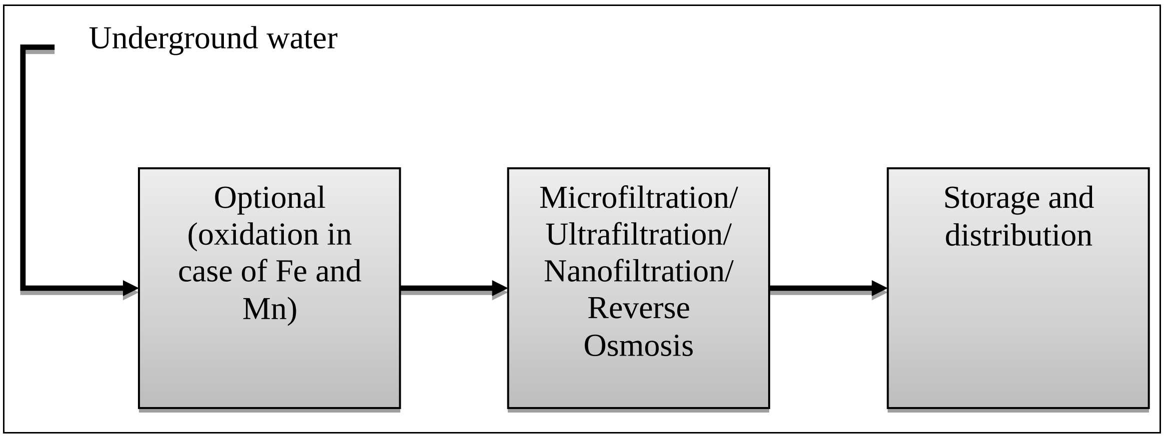

Figure 2-7 Integrated membrane system for clean water (Brikkke and Bredero, 2003)

2.3.7 Conventional clarification pre-treatment

Conventional clarification used as pre-treatment to NF/RO is mostly used when upgrading from an existing conventional water treatment facility to IMS. In such processes it is the chemical disinfection that is replaced by NF/RO. A study by Glueckstern and Priel (2002) revealed that conventional clarification is used as pre-treatment in cases where there is little experience with MF and UF systems and also low capital investment projects. The major limitation of the conventional clarification is its inability to cope with fluctuations of the incoming raw feed quality. The poorly clarified effluent from conventional clarification is passed to the NF/RO processes which results in its fouling and high operating cost (Glueckstern and Priel, 2002; Radcliff and Zarnadze, 2004; Knops *et al.,* 2007). The study by Zhou *et al.* (2009) shows that some of the coagulants used in conventional clarification contributed highly to fouling in the NF/RO membranes. This could happen if they dissolve and end up on NF/RO membranes.

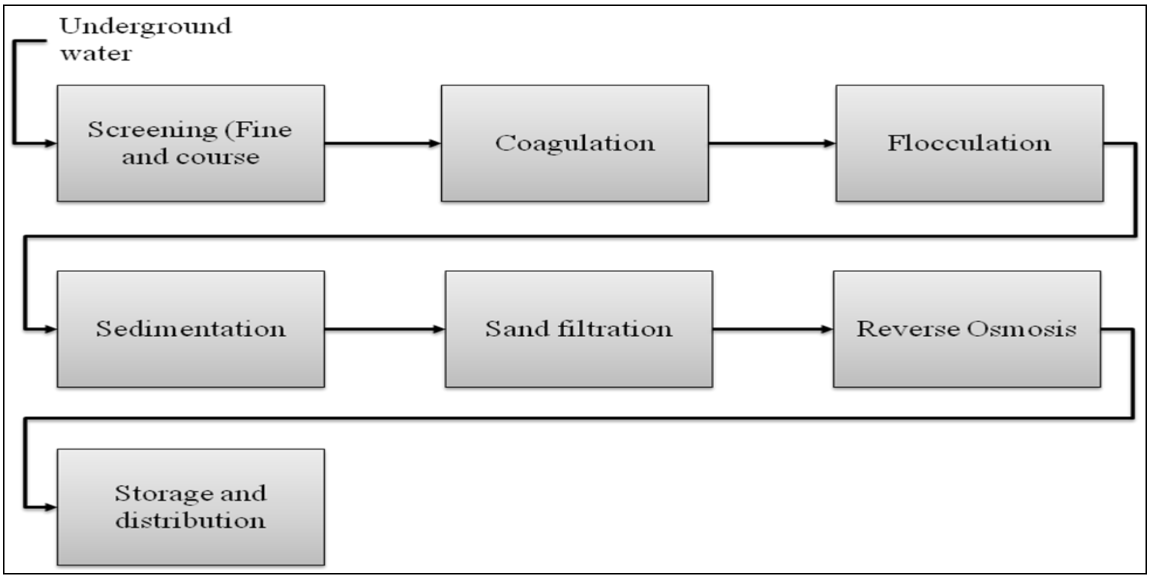

Figure 2-8 Integrated membrane system with conventional clarification set-up (Cheremisinoff, 2002)

2.3.8 Microfiltration/Ultrafiltration pre-treatment

MF/UF processes are increasingly being used as pre-treatment processes for NF/RO in desalination, wastewatertreatment and in most IMS systems for drinking water treatment. The factors favouring the use of MF/UF as pre-treatment for NF/RO are, a need for high consistent quality from upstream processes, small foot print and reduction/elimination of chemicals used in the upstream processes (Von Gottberg *et al.*, 2005; AwwaRF, 2006; Karlapudi *et al.*, 2012). Choosing between MF and UF is determined by the fouling propensities of the feed water. UF membranes are less susceptible to fouling by colloidal matter than MF membranes, but are more prone to macromolecular fouling/biofouling than MF membranes. In terms of protecting NF/RO membranes from dispersed particles, UF offers more protection than MF or conventional clarification (Schäfer *et al*,. 2000).

2.3.9 Process description for MF/UF pre-treatment

Raw water from the storage is pumped to the first filtration process which is usually MF/UF (low pressure membrane system). Depending on the filtration process used, normally all the dispersed particles (suspended solid, colloidal matter, bacteria and some viruses) are removed in the first filtration process. The filtrate from the low pressure membrane filtration process is pumped to a high pressure membrane filtration process which is either NF/RO. The remainder of the viruses, bacteria, dissolved organic and inorganic materials are removed by the high pressure membranes. Virus and bacteria removal is 3-6 log in the low pressure membrane system and is about 8 log in the high pressure system (Zhou *et al.*, 2009). Post NF/RO, chemical addition is usually required for water stabilization, pH adjustment and chlorine residual. The latter is dependent on the regulatory board (Bilqis *et al.*, 2006).

2.4 Hydraulic configuration

Membrane processes can be operated in different configurations namely; deposition mode, suspended mode and semi-suspended mode. Each of the configurations comes with its own operational methodology and limitations.

2.4.1 Deposition mode

The deposition mode is commonly known as the dead-end mode. In the dead-end mode, the entire mixture to be filtered is perpendicularly forced through the membrane by pressure (Fig 2-9). The particles bigger or equal to the membrane pore size are trapped and retained on the membrane surface. Whilst the pure water/solvent passes freely through the membrane pores. As the filtration time increases the retained matter on the membrane surface or pore channel increases thereby increasing the required pressure to continue to drive the fluid through the membrane (Baker, 2004).

In dead-end operation, the frequency of stopping to clean the deposited layer of particles on the membrane surface is high. A feed with high contamination levels requires frequent cleaning due to increased load (Pilutti *et al.*, 2003). In dead mode operation there is only one influent stream (feed) and one effluent stream (filtrate), the contaminants are held in place by driving pressure on the feed side of the membrane (Allgeier, 2005). According to Baker (2004) dead-end mode is more suitable for depth filtration, as is the case with conventional sand filtration.

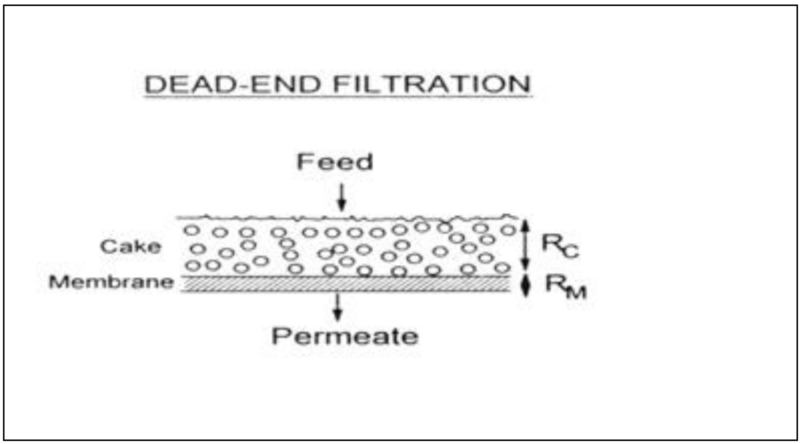

Figure 2-9 Schematic drawing for dead-end/deposition mode (Cheryan, 1998)

2.4.2 Suspended mode

Suspension mode is commonly known as cross-flow mode, where the feed flows parallel to the membrane. The parallel flow to the membrane surface is used to disperse the accumulated layer that develops on the membrane surface due to the filtration rejection process. Dispersing the accumulated layer minimizes the accumulation of particles on the membrane surface, resulting in high fluxes and lower pressure drop across the membrane. Dispersing of the accumulated matter on the membrane surface is done by creating turbulence by the high velocity of the fluid at the membrane surface.

Turbulence created prevents solution stagnation on the membrane surface (Allgeier, 2005). Figure 2-10 shows the schematic drawing for cross-flow operation or suspended mode of operation. Suspended mode is only used in surface/screen filters where the accumulation of matter is on the surface of the membrane and not inside the pore, as it is the case with depth filters (Baker, 2004). Other means of creating turbulence on the membrane surface is by suspended carriers and air. The cross-flow mode reduces the need to stop and clean the membrane either by backwashing, pulsation or any other cleaning method. Cross flow mode is the preferred mode in the industry compared to the dead end mode (Sim *et al.,* 2011).

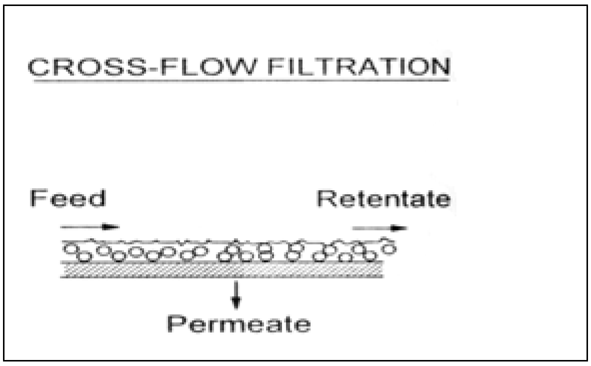

Figure 2-10 Schematic drawing for cross flow operation/suspended mode (Cheryan, 1998)

2.4.3 Semi suspended mode

Semi-suspended mode is the transition between completely dead end and cross flow mode. The submerged membrane configuration falls under the semi suspended mode. Submerged membrane configuration has wide applications, especially in membrane bioreactors (MBR). Figure 2-11 shows a schematic drawing for immersed membrane operation or semi suspended mode. In this mode a third medium such as air (commercial) or beads (laboratory/pilot plant) is used to create turbulence on the membrane surface and disperse the accumulating matter on the membrane surface. In MBRs, a membrane pack is immersed in the liquid which needs to be separated. The driving force can either be gravity or pump suction. In a gravity driven process the liquid head drives the filtration process. The driving pressure is applied on the feed solution being separated. While under pump suction, the driving force is the negative pressure created by the pump suction connected to the permeate side of the membrane pack. An air diffuser is connected at the bottom of the membrane pack through which air bubbles are supplied to create turbulence at the membrane surface and prevent accumulation of particles on the membrane surface (Shino et al., 2004). Other ways of changing the process from a deposition mode to the suspended mode configuration is by frequent backwashing, frequent pulsation or a combination of all (Cheryan, 1998).

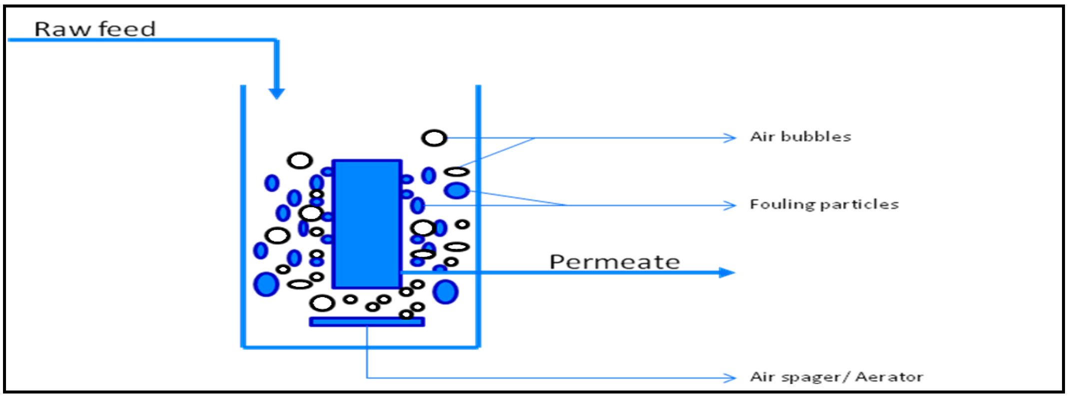

Figure 2-11 Schematic drawing for immersed membrane operation/suspended mode modified from (Cheryan, 1998)

Table 2-3 Summary of the comparison of membrane operation mode and typical uses

Configuration	**Advantages**	**Disadvantages**	**Applicability**
Submerged without scouring	Good rejection (improves with time), reduced foot-print	Severe fouling leading to difficulty in cleaning, batch operation	Rural water treatment/ low drinking water production
Submerged with scouring	Continuous process, steady fluxes, reduced fouling, reduced foot-print	Energy for scouring effect	Wastewatertreatment (MBR)
Cross-flow	Continuous process, steady fluxes, reduced fouling	Pumping cost, wide area required	Urban drinking water production, industrial water treatment
Dead-end	Good rejection (improves with time), reduced foot-print	Severe fouling leading to difficulty in cleaning, batch operation	Analytical application, cell harvesting (MBR or pharmaceuticals)

2.5 Membrane modules

Application of membranes on an industrial scale requires a large membrane area, packed in a fairly small fractional area if the technology is to be economically viable and meet production demands. Membrane manufacturers are therefore required to economically and efficiently package membrane modules into manageable areas before they can be used in an industrial application (Baker, 2004). A membrane module is a piece of equipment in a filtration unit in which a membrane area is packaged. The module must be easily removed and replaced and preferably offer some means to trouble-shoot or isolate defective membranes (Noble and Stern, 1999). A membrane medium itself is manufactured either as a flat-sheet stock or hollow-fibre, it is only configured later into different modules. The membrane module process involves potting or sealing the membrane material into a specific assembly, which may incorporate an integral containment structure of the filtration unit (Allgeier, 2005). Typical membrane modules are hollow-fibre, spiral wound, tubular and plate and frame. Module design should be able to deal with the following issues; economy of manufacturing, membrane integrity against damage and leak, sufficient mass transfer to keep polarization in control, minimise waste of energy, easy to allow permeate to pass through and should allow the membranes to be cleaned.

2.5.1 Tubular modules

Tubular modules are amongst the earliest commercial industrial membrane modules designed. Figure 2-12 shows typical examples of tubular membrane modules. The design consists of a tubular pressure tube(s) and tubular membrane. The membrane is inserted or applied inside the pressure tube(s) which serves as a support for the membrane. The pressure tube(s) are perforated or permeable to permeate. Feed is pumped through these tubes and is collected in the outside space between the pressure tube and supporting tubes and is then withdrawn at a connecting piece on a pressure tube. The permeate is collected on the shell side of the tubes (Pinnekamp, 2003). The supporting tubes are typically made of stainless steel with an internal diameter of 12.5 mm and an external diameter of 14 mm. Due to the large diameter of the tubes, these modules are able to handle feeds with large particles i.e. slurries. Tubular membranes operate in turbulent region at velocities between 2 to 6 m/s. The high velocity is primarily for fouling control. It is easy to clean these membranes *in situ* and individual tubes can be separated if malfunctioning. The biggest disadvantage of tubular modules is the low surface area-to-volume ratio; it has the lowest of all module configurations (Cheryan, 1998).

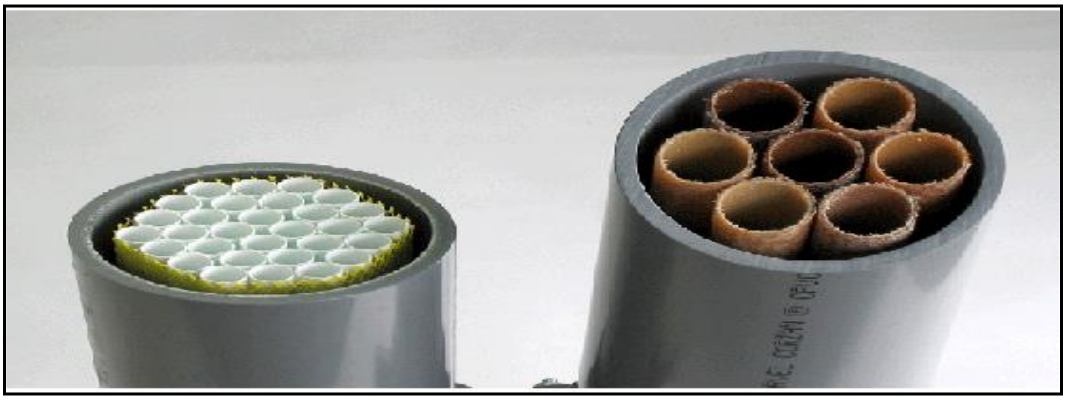

Figure 2-12 Tubular membrane module presentation (Yoon, 2011)

2.5.2 Hollow-fibre modules

Hollow-fibre module configuration is similar to that of tubular modules. The major difference is that the membranes in hollow-fibre modules are in a self-supporting tube with no separate support or backing. The feed could either be on the inner tube side or shell (outer). Prior to MBR systems most of the MF and UF hollow-fibre modules had the feed on the inner side of the tube. Only RO systems had the feed on the shell side. The fibres come in different diameters ranging from 0.2 to 2 mm. The thickness ranges from 100 to 400 µm and a packing density for hollow-fibre modules ranges from 50 to 3000 of individual fibres. The fibres are sealed into a hydraulically symmetrical housing in a shell-and-tube arrangement and bonded on each end with epoxy. The housing material can be of any material, but the most common are, polyvinyl chloride (PVC) and stainless steel.

The length of the fibre is decided by the application. If dilute, non-fouling operations that are pressure dependent but flow regime independent, then long fibers are used. The opposite is true for viscous and fouling processes. Hollow-fiber membrane modules are operated in a laminar region, at low pressure drops and moderate flow rates. These modules have the highest surface area to volume ratio out of all the module configurations. They are susceptible to particle plugging hence pretreatment is necessary. Their self-supporting nature makes them versatile for most of the fouling control strategies. This is their biggest advantage. The worst experience with hollow fiber modules is the high replacement cost (one leaking tube can lead to a replacement of the entire module and identification of the leaking module is tedious) (Cheryan, 1998). Figure 2-13 shows some hollow fibre-modules.

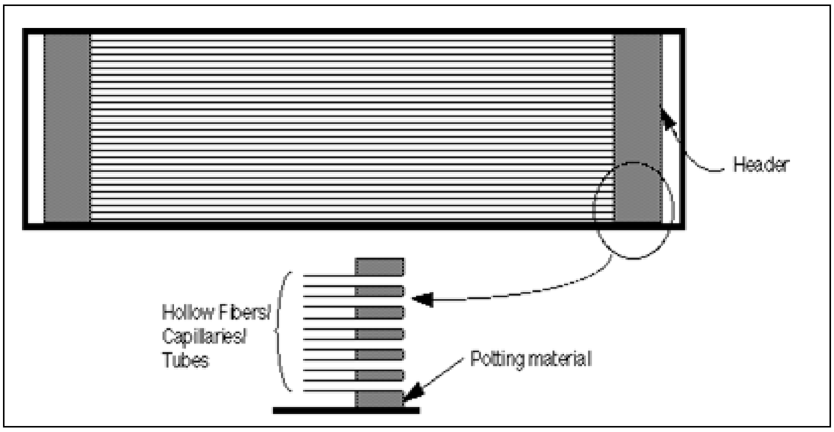

Figure 2-13 Hollow-fibre presentation (Baker, 2004)

2.5.3 Spiral-wound modules

Spiral-wound modules were introduced first as an efficient configuration for mostly NF/RO modules. The form of a spiral wound module is a sandwich arrangement of a flat-sheet membrane wounded around a central perforated tube. The sandwich is made up of two sheets of membranes placed back to back, and separated by a spacer which is used to channel permeate to its exit. The sheets are glued on three sides and the unglued side is sealed around the perforated centre tube. Feed water enters the spacer channels at the end of the spiral-wound element in a path parallel to the central tube. The filtered water in the permeate carrier travels spirally inward around the permeate carrier towards the central collector tube, while the water in the feed spacer that does not permeate through the membrane layer continues to flow across the membrane surface (Allgeier, 2005).

Several layers of the membrane sandwiches are placed on top of each other and rolled together. A finished spiral module is placed inside a PVC or stainless steel housing fitted with the suiting end-caps and manifolds for the filtration process (Cheryan, 1998). According to Cheryan (1998), spiral modules operate in a turbulent region. The turbulence is created by the spacer in-between the membranes. Spiral modules are operated at high pressure drops and the thickness of the spacer affects the pressure drop. Advantages of spiral wound modules include a good packing density, easy to remove and replace and has the lowest capital cost amongst all the module designs. The disadvantage of spiral-wound is that although replacement is easy, the design on collection points (both permeate and retentate) are different for each manufacture. This then means modules cannot be changed across companies without affecting the process design. Another disadvantage is that large spacers trap-in particles while small spacers limit the flow rate through the module, making it only good for clean, non-viscous and non-fouling fluids (Cheryan, 1998).

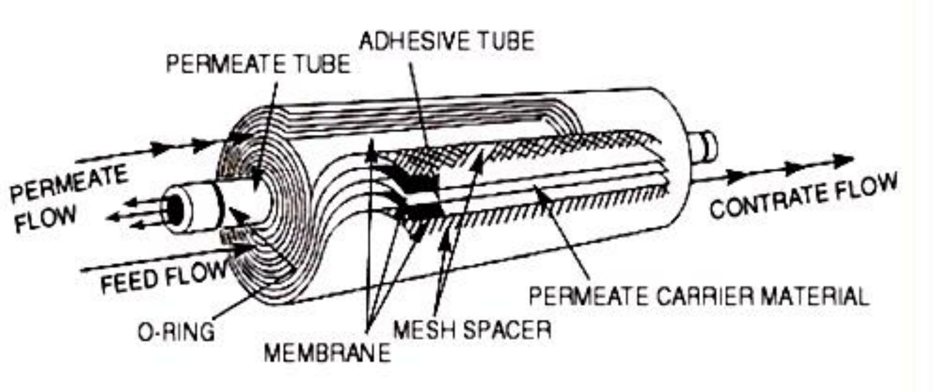

Figure 2-14 Spiral wound module presentation (Baker, 2004)

2.5.4 Plate and frame modules

Plate and frame modules are also amongst the first modules designed for industrial use (Cheryan, 1998; Baker, 2004). A plate membrane module consists of two sheets of membrane materials stuck/glued together on four sides/edges. In between the two sheets, there is a spacer/mesh to channel permeate and prevent the two sheets from getting stuck together during filtration and stopping the permeate flow. The permeate outlet nozzle is placed on one of the four sides of the plate membrane module. Plate membrane modules are usually stacked on top of each other to form a frame.

Figure 2-15 shows some flat-sheet membrane modules from Kubota. A single plate membrane module has a typical area of 0.1 m^2 to 0.4 m^2. In between the adjacent plate membrane module is a frame and a space for feed flow/fouling control. Each plate membrane module in a frame connects to a single manifold which receives the permeate from each plate membrane module via tubes. The single manifold is usually situated peripheral to the process (Baker, 2004). Plate and frame modules are typically operated in a submerged configuration. Advantages of the plate and frame modules are that they are easy to remove and replace, are moderate in capital cost and easy to troubleshoot or even isolate a suspected leaking membrane module. Operation of plate and frame modules is in a laminar region, hence these are susceptible to fouling (Cheryan, 1998).

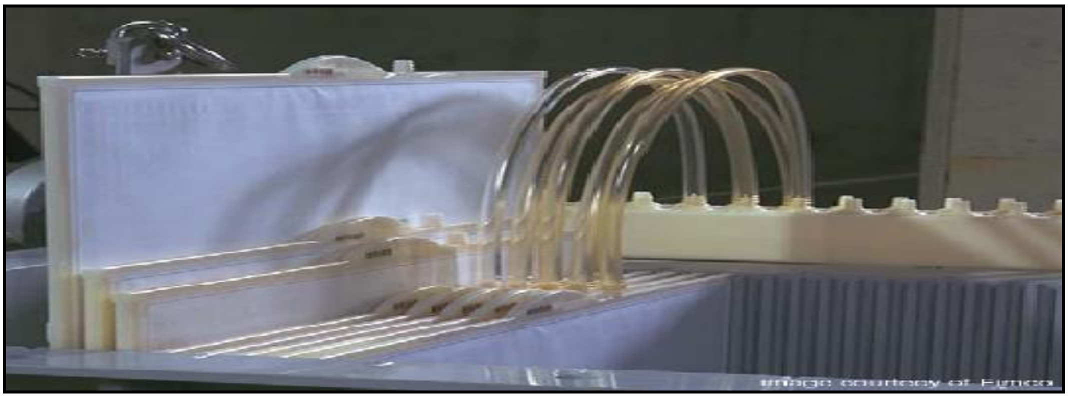

Figure 2-15 Flat sheet module presentation (Yoon, 2011)

2.6 Membrane material and transport

Membrane material varies from one to the other. The most important consideration made in choosing the material to be used is that it must provide a separation performance that conforms to what a membrane does. This is the reason why a variety of membrane materials are commercially used (Noble and Stern, 1995). A factor that is now increasingly being considered in membrane materials is the material’s susceptibility to fouling. Hydrophilic membranes have higher fouling resistance than hydrophobic membranes (Chan and Chen, 2004). This means that hydrophobic membranes can be impregnated with water soluble materials such as poly-vinyl pyrrolidone or poly-vinyl methyl ether. This is mostly at the polymer formulation stage. The charge of the membrane material also affects fouling particularly in water treatment. For example, colloidal matter is slightly negatively charged due to carboxyl and acidic groups. In a situation where the same charge ions exist between the membrane and the colloidal matter, the fouling effect is reduced resulting in high flux due to repulsive forces between the membrane and the colloidal matter. The opposite is true for oppositely charged membranes and colloidal matter (Baker, 2004). Other important parameters considered when selecting membrane material is the process hydraulics. Most membrane material fails to be commercialized because of their weak tensile strength, while others fail due to their chemical strength. This is because all membranes during a filtration process will at some stage require some chemical cleaning (Cheryan, 1998). Table 2-4 gives a list of some membrane materials.

Table 2-4 List of the most used commercial material for manufacturing of MF (Cheryan, 1998)

Hydrophobic materials	Polyletrafluoroethylene (PTFE Teflon)
	Poly-vinydene fluorid (PVDF)
	Polyproylen (PP)
	Polyethylene (PE)
	Polyester
Hydrophilic materials	Cellulose ester
	Polycarbonate (PC)
	Polysulfone (PSF)
	Polyimide (PI)
	Aliphatic polyamde (AP)
	Poly Etherimide (PEI)
	Poly Ethersulfone (PES)
	Polyetheretherketone
Ceramics	Alumina (Al_2O_3)
	Zicania (ZiO)
	Tilania (TiO_2)
	Silicium Cabide (SiC)

2.6.1 Membrane flux

Membrane flux is the amount of volumetric flow rate through a filtration/membrane area per unit time. The general unit for flux is litres per square metre area per time in hours (LMH). Flux is affected mainly by four factors; pressure, concentration, temperature and turbulence adjacent to membrane surface. The driving force for most commercialized membrane systems is pressure. The relationship between pressure and flux is positive. A rise in pressure causes a rise in flux; however, for feed mixtures there is a point where further increases in pressure results in minimal increase in flux. This is because the particles of the component being rejected by the membrane accumulates on the membrane surface and obstructs the passage of the solvent through the membrane. If the process is allowed to continue to run, the rejected layer on the membrane surface grows thicker and becomes more and more resistant to solvent flow and this result in the flux dropping. At this point it is said that the membrane is fouled and it is no longer economically justifiable to continue to run because the added energy to the system does not recover or even maintain flux (Miller *et al.,* 2013).

Other factors affecting flux are feed concentration, feed temperature and membrane pores. High concentration of the rejected particulates in the feed mixture results in a quicker build-up of the layer that obstruct solvent flow as it also affects viscosity. Temperature affects mainly the density and viscosity mixture. Dense and viscous liquids tend to form a gel layer on the membrane surface that is highly resistant to permeation. Tight membranes or a membrane with small pores have low permeability because such a membrane will reject more particles and it is easy for the gel layer to form on tight membranes. Turbulence at the membrane surface reduces the concentration polarization layer at the boundary, and improves back diffusion close to the membrane surface which improves mass transfer and in turn improves flux (Cheryan, 1998).

2.6.2 Transport through a membrane

Filtration in the membrane process is driven either by pressure or mass transfer. Membrane technology has been in existence for over two and half decades but there has been minimal success to come up with a universal model to predict flux for MF and UF, due to several changes in the physical properties and hydraulics during a singular filtration cycle.

(i) Pressure- Flux model

Flow or flux through a membrane is described by the Hagen-Poiseuille equation for cylindrical pores, typically used with MF and UF. Hagen-Poiseuille’s equation is as follows:

$$J = \frac{\varepsilon . d_p{}^2 P_T}{32 \Delta x \mu} \qquad \textbf{2- 6}$$

J is the flow rate through the membrane, measured in LMH; d_P is the channel diameter (m), in this case the mean pore diameter; P_T is the applied transmembrane pressure; μ is the viscosity of the fluid permeating the membrane (Kg/m.s); Δx is the length of the channel, the membrane "skin" thickness (m); and ε is the surface porosity of the membrane.

The driving force in equation 2-6 is the applied pressure, and flux is a direct function of this pressure. The effect of pressure remains only true if the surface of the membrane is not altered in the process of filtration, by either particle deposition on the membrane surface/pore or concentration polarization which is a function of the concentration of solutes in the feed mixture. In a filtration process it is almost impossible to stay within the constraints of the pressure flux model because of the nature of the process, which is to concentrate the feed side, hence, causing changes on the membrane surface. To attempt to remain in this ideal condition, turbulence, close to the membrane surface is to be created to disperse the developing layer of solutes on the membrane surface. If the layer of solutes on the membrane surface is allowed to grow, flux becomes independent of pressure and this phenomenon is common in membrane filtration processes. Also, the viscosity of the rejected solutes changes as the concentration on the membrane surface changes. The rejected particles block some of the pores and the thickness of the cake layer on the membrane surface also grows. Because almost all of the parameters in the Hagen-Poiseulle’s equation change as the filtration process progresses, prediction of flux using this equation is not possible (Cheryn, 1998).

(ii) Mass transfer model

During microfiltration or ultrafiltration, solutes in the feed are brought to the membrane surface by convection transport. The rate of transporting solutes to the membrane surface is a function of filtration flux and solute concentration.

$$J_s = JC_B \qquad \textbf{2-7}$$

J_s is convection transport rate of solute to the membrane surface (m/s) J permeate rate through the membrane (m/s) C_B is the bulk concentration of the rejected solute (kg/m^3).

The resulting concentration gradient on the membrane surface or close to the membrane causes the solute to be transported back into the bulk solution due to diffusion effects. The rate of the back diffusion is defined by the following equation;

$$J_s = D\frac{dC}{dx} \qquad \textbf{2-8}$$

D is the solute molecular diffusivity (m^2/s) and dC/dx is the concentration gradient over a differential element in the boundary layer. At steady state, the two mechanisms will balance each other.

Combining equation 2-7 with 2-8 a mass transfer equation is derived and is as follows;

$$J = \frac{D}{\delta}\ln\frac{C_G}{C_B} \qquad \textbf{2-9}$$

δ is the thickness of the boundary layer over which the concentration gradient exists**,** C_G is the gel concentration at the membrane surface, C_B is the bulk concentration.

The mass transfer model is only applicable in a pressure independent application. Flux at this stage is controlled by the rate of solute diffusion away from the membrane surface to the bulk solution. Generally, C_B and C_G are determined by physicochemical properties of the feed. Flux control in mass transfer operation is achieved by controlling the boundary layer thickness. When this layer is reduced, as can be seen in equation 2-9, the net result is higher permeation flux through the membrane. Controlling or reducing the thickness of the boundary layer at the membrane surface improves flux both under mass transfer operation and under pressure-flux operation. Improving hydrodynamic conditions around the membrane surface improves flux produced under the mass transfer operation. Because improved hydrodynamic conditions creates turbulence on the membrane surface, the thickness of the boundary layer is reduced which in turn results in higher flux. However, turbulence at the membrane surface reduces C_G which ends up reducing flux. The benefit of turbulence over high C_G is that flux produced under mass transfer operation is lower in relation to that produced under pressure-flux operation. High C_G under pressure-flux operation results in lower flux due to increased osmotic pressure on the membrane surface and increased hydraulic resistance (Van de Waal and Racz, 1989).

(iii) Resistant model

Both the mass transfer model and pressure flux model do not explain the flux in high pressure filtration operations. Mass transfer is more appropriate for low pressure operations while pressure flux model is appropriate for medium pressure operations. Operations at high pressure can be explained by the resistance in series model (Cheryan, 1998):

$$J = \frac{P_T}{R'_M + R_G} \qquad \textbf{2-10}$$

R'_M is a combination of membrane resistance of the clean membrane (R_M) and fouling (R_F) resistance

$$R'_M = R_M + R_F \qquad \textbf{2-11}$$

R_G is a function of operating parameters and physical properties. These parameters are permeability of the gel and its thickness (boundary layer on the membrane surface), which is a function of applied pressure.

$$R_G = \phi P_T \qquad \textbf{2-12}$$

R_F signifies membrane fouling which is formed because of specific membrane-solute interactions. The intrinsic membrane resistance may change. The ϕ term is a function of the variables affecting mass transfer such as viscosity, shear rate/velocity, and temperature. With the resistance model as well, control of the thickness of the boundary layer at the membrane surface improves the flux through the membrane.

(iv) Osmotic flux model

The gel layer that forms on the surface of the membrane whenever a real feed is filtered in a membrane has some resistance to flux and this resistance is termed osmotic resistance and it affects the required driving pressure to achieve filtration.

$$J = \frac{P_T - \Delta\pi}{R_M} \qquad \textbf{2- 13}$$

$\Delta\pi$ is osmotic pressure and is determined only by C_B

2.7 Membrane properties

Membrane properties must be discussed together with foulant type because of the interaction that takes place between the two during filtration. The major properties of the membrane that affect fouling are; hydrophobicity, charge, roughness and pore size.

The general approach in the minimization of membrane fouling when the fouling characteristics of the feed are known is by selecting a membrane which has low fouling affinity of the feed solution (Nguyen *et al.,* 2012). The hydrophobicity of the membrane material plays a major role in membrane fouling. Hydrophobic interaction can be described as “like attracts like” that is, there is a natural tendency of attraction between membranes and solutes with similar chemical structures. Hydrophobic attraction results from van der Waals forces between molecules (Abdelrasoul *et al.,* 2013). Hydrophobic adhesion is an important mechanism for fouling dominated by NOM, because high molecular weight NOM provides great potential for hydrophobic adhesion because of their charge density. Other factors that affect the strength of the adhesion to membrane surfaces are membrane surface roughness and membrane pore size (Flemming and Schaule 1988; Liu *et al.* 2000). A study by Bendinger *et al.* (1993) showed that most foulants that are hydrophobic and slightly hydrophilic adhere better on hydrophobic surfaces than on hydrophilic surfaces. Only highly hydrophilic foulants attach stronger on hydrophilic material. Extremely hydrophobic materials did not adhere too well on the hydrophobic and hydrophilic material.

There has been many more studies that have confirmed Bendinger *et al.* (1993) findings that hydrophobic membranes are more prone to fouling than hydrophilic membranes (Chan and Chen, 2004; AwwaRF, 2008; Abdelrasoul *et al.*, 2013). Bessiere *et al.* (2009) showed that a hydrophilic membrane was fouled more by the hydrophilic components in the feed mixture than hydrophobic components. This is also in agreement with the study by Bendinger *et al.* (1993). However, a study by Lee *et al.* (2004) showed no difference in fouling of hydrophobic or hydrophilic membranes by five different rivers. The parameters that affected fouling were membrane surface roughness and membrane pore size. The rough and large pore membranes were shown to be more prone to fouling than the smooth and small pore membranes. This is because the bigger pores are more accessible to foulants. Nguyen *et al.* (2012) found that rough surfaces have areas where there is stagnation of liquid or laminar flow due to trenches on the membrane surface. In the trenches (uneven surfaces) the biofilm begins to develop and binds strongly to the surface of the membrane. A study by Hashino *et al.* (2011) showed that fouling by biofilm and colloidal matter was found more in the trenches of the membrane than on the even surface.

2.7.1 Hydrodynamic conditions

Hydrodynamic conditions in membrane technology mostly refers to initial permeate flux, transmembrane pressure and turbulence at the membrane surface. Hydrodynamic condition controls the rate of particle deposition on the membrane. Initial flux is a term used mostly with constant pressure systems, initial flux is the flux at the beginning of filtration and is usually high because at this stage the membrane is clean. Due to high initial flux, particles in a suspension are dragged towards the membrane surface faster than they are diffused/dispersed back into the bulk solution. Therefore, more particles are deposited on the membrane during high initial flux (Yoon, 2011). The study by Hwang *et al.* (2008) showed that under high initial flux a large number of particles are simultaneously transported towards the membrane surface. The simultaneous arrival of these particles on the membrane surface was found to be the factor that benefits flux because entry to the membrane pore is hindered and particles only deposit on the membrane surface rather than the membrane internals. The opposite was found for a low initial flux. The finding of Hwang *et al.* (2008) was also confirmed by Wang and Tarabara (2008).

Particle deposition rate or fouling of a membrane is proportional to flux and or transmembrane pressure (TMP) and flux is proportional to TMP in a constant flux operation. since TMP affects both the fouling rate and flux, the critical flux concept was developed. At critical flux only localized deposition on the membrane occurs, because the rate at which particles deposit on the membrane surface is almost equal to the rate at which they are diffused back into the solution (Bacchin, 2004). However, if the process is operated above the critical flux catastrophic fouling is observed on the membrane because of increased deposition on the membrane surface and the additional TMP compresses the cake layer making it hydraulic resistant to flux (Cheryan, 1998). The study by Grandison *et al.* (2000) showed that increasing TMP increases both the reversible and irreversible fouling when the other parameters that hinder deposition are kept constant. Mosqueda-Jimenez *et al.* (2008) found that high fluxes/TMP cause irreversible fouling on the membrane due to operation above that critical flux of the system. The cake layer formed during this study was found to be compact and highly resistant to permeation. Increasing TMP or flux which increases the drag force that pulls particles towards the membrane requires that the cross-flow velocity/turbulence at the membrane surface to be increased in order to avoid excessive fouling (Kumar *et al.*, 2007).

In membrane filtration, rejected particles by the membrane accumulate near the membrane surface due to hindered effects. According to Eckstein *et al.* (1974) particles in a close proximity to each other collide with each other more frequently than those at far distance to each other. This study showed that if the velocity of the liquid is increased, the collisions are more vigorous. The collision between particles results in high diffusion of particles back to or away from the membrane surface hence increasing flux. Cross-flow velocity and air scouring is mostly aimed at improving shear induced diffusivity as well as control/removal of the cake layer on the membrane surface. In tubular modules and spiral modules cross-flow velocity is achieved by increasing feed fluid velocity or re-circulating rate (Cheryan, 1998). In submerged membrane systems the turbulence on the membrane surface is achieved by air scouring where the rising air bubbles on the exterior of the membrane create the shearing effect on the membrane. When the rate of scouring is increased the fouling rate or development on the membrane is reduced, but there is a limit to which the improvement is possible (Yamamoto *et al.*, 1989; Menniti and Morgenroth, 2010). Air scouring is more energy efficient than the liquid cross-flow filtration and does not break the flocs as with tradition cross flow filtration which in turn results in severe fouling (Berube *et al.*, 2006). According to the membrane filtration theory, at sub critical flux there is no deposition of particles on the membrane surface. Therefore, in order to keep membrane filtration at sub-critical flux, if flux is increased, the cross flow velocity or scouring rate must also be increased (Delgado *et al.* 2008). Other means of creating or improving particle collision on the membrane surface, particularly in laminar conditions, is by pulsation. This is achieved by pressurizing and depressurizing of the permeate side and this method was found to be effective in controlling fouling on the membrane surface (Hadzismajlovic and Bertram, 1998).

2.8 Fouling

Fouling is an irreversible alteration of a membrane due to specific physical and or chemical interactions between the membrane and various components present in the process stream. Membrane fouling manifests as a decline in filtrate flux and a change in membrane selectivity. The irreversible changes on the membrane that is due to fouling, continues throughout the process until extensive cleaning or replacement of the membrane is done (Ho and Sirkar, 1992).

An inherent behaviour of any filtration system is the accumulation of solutes or particles on the membrane surface as they get rejected by the membrane. These accumulated material results in a reduction in permeate flow, but the reduction in permeation is reversible when the driving force is removed or permeation is stopped (Field *et al.,* 2010). This is only true in cases where the dominant foulants in a solution are macromolecular solutes which form a gel layer on the membrane surface. At this stage of filtration the reduction in flux is believed to be due to concentration polarization. However, if the driving force (transmembrane pressure) is increased further in an attempt to increase flux, only a momentary increase is observed which quickly declines. This act causes the gel layer thickness to grow and as it grows its resistance to filtration increases. Any further increase in transmembrane pressure from this point causes fouling to start developing on the membrane layer. The high pressure is mostly used up in compressing the layer of foulants that is already attached on the membrane surface rather than improving flux (Bacchin *et al.*, 2002).

In a solution containing colloidal material, fouling begins when the driving force/permeation rate moves the process from sub-critical flux to critical. Critical flux is a criterion for the transition between concentration polarization and fouling. At critical flux, localized irreversible fouling occurs on the membrane. If the driving force is increased beyond this point, limiting flux occurs, where the entire membrane operates at critical flux. Critical flux is dependent on several parameters which include suspension of properties and hydrodynamic conditions. Hydrodynamic conditions are a function of tangential velocity (Bacchin, 2004). At critical flux, the rate at which the fouling particles are dragged and deposited on the membrane surface is greater than the rate at which the particles are dispersed away from the membrane surface. Once the particles have deposited on the membrane, increasing transmembrane pressure simply adds another layer of fouling on the membrane and fouling becomes more severe (Bacchin *et al.*, 2002).

Membrane fouling still continues to be a concern, five decades after the application of the technology (Tarragona, 2005). The challenge is that the entire components in a process stream being separated can foul a membrane. The nature and the extent of the fouling are determined by the specific physical and chemical characteristics of the individual components and the membrane (Abdelrasoul *et al.*, 2013). In many systems, the species causing most of the fouling are present in only trace amounts and their concentrations may be virtually independent of the main components involved in the actual separation (Zeman and Zydney, 1996).

The major challenge in bringing about a fundamental understanding of membrane fouling is the difficulty in identifying clearly the actual foulants, and distinguishing between the symptoms of fouling, effects of concentration polarization and membrane compaction. Many researchers have mistakenly attributed rapid decline in flux seen during the initial stages of filtration process to concentration polarization and the long term decline after the initial sharp decline to fouling. While in actual fact fouling can occur just as rapid, for example protein adsorption or solid substrate on the membrane surface (Zeman and Zydney, 1996).

Membrane fouling is managed under three main headings, feed composition, hydrodynamic conditions and membrane properties. Proper management of these can make the filtration process economical and are managed at different stages (Tang, 2011).

2.8.1 Effect of concentration on fouling

Suspension properties play a major role in membrane fouling. These properties include foulant type, particle size and concentration. Influence of feed concentration on membrane fouling as investigated by Huang *et al.,* (2013) has greater impact than transmembrane pressure. This study shows that if the foulant concentration in the feed is increased towards its critical point, the fouling propensity of the feed is high and the transmembrane pressure has very little influence on flux. From the mass transfer model, (equation 2-9), it is seen that if the bulk/feed concentration is close to the concentration on the membrane surface, no back diffusion is possible. The driving force (transmembrane pressure) acts to compress the layer on the membrane surface, making it more resistant to permeation (Zydney and Colton, 1986). Feed with high concentration of the particulates to be separated by the membrane has higher fouling tendencies on the membrane because the particulates accumulate on the membrane surface. The accumulated particulates obstructs and resist the permeate flow from the membrane (Bessiere *et al.,* 2009).

2.8.2 Effect of foulant type and particle size on fouling

(i) Natural Organic Matter (NOM)

Natural organic matters (NOM) are known as allochthonous. NOMs which are terrestrially derived are known as autochthonous NOM. These are also the microbially derived and wastewater NOM. Each of these organic fractions fouls membranes differently because of different hydrophobicity, molecular weight size and charge density. However, microbially derived NOM are found to be the worst foulants. Fouling from these fractions is found to be most problematic and severe (Zhou *et al.*, 2009). A study by Bessiere *et al.* (2009) on the effect of NOM on fouling shows that hydrophilic components of NOM are responsible for the rapid but reversible fouling on the membrane. The hydrophobic components were found to be responsible for the slow but irreversible fouling on the membrane. The hydrophilic components were small compared to the hydrophobic components. Both of these components get adsorbed on the membrane material.

The size of NOM plays an important role on the fouling of the membrane. Because NOM adsorbs onto the membrane, small NOM enter the pore of the membrane and get adsorbed on the wall of the pore channel resulting in pore narrowing. Larger NOM components get trapped at the entrance of the membrane pores and blocks the entrance to the pore channel resulting in cake layer formation as filtration progresses (Kim *et al.*, 2006). Fan *et al.* (2014) found that the fouling order of hydrophobic membranes by NOM material is as follows; hydrophilic neutrals > hydrophobic acids > transphilic acids.

Colloidal matter in a suspension can be charged and depending on the charge of both the membrane and the particle, adhesion or repulsion will occur. The charge of the particles can be altered by adjusting the pH of the suspension. pH adjustment changes the electrostatic interaction between the membrane and particle from attractive to repulsive or from repulsive to attractive (Madaeni, 1999). The effect of the ionic strength of colloidal particles on fouling was also studied by Singh and Song (2005). The study found that increasing the ionic strength of colloidal matter and its concentration in the feed solution increases the fouling potential of the water linearly.

(ii) Fouling by Inorganic Compounds

Inorganic fouling normally results from the accumulation of inorganic matter/precipitates such as metal hydroxides and silica on the surface of the membrane. Inorganic fouling will foul the membrane both on the surface and internally. The precipitates are formed when the concentration of the chemical species exceeds their saturation concentrations. This tends to happen on the membrane surface where accumulations of particles occur due to particles being retained by the membrane. Inorganic fouling is more common with NF and RO but it is also found in MF and UF. With MF and UF, it mostly exists due to interactions between ions and other fouling materials such as organic polymers. The effects of inorganic fouling in MF and UF are not as severe as it is in NF and RO membranes (Liu *et al.,* 2000).

(iii) Protein Fouling

There is a very wide range of proteins and there is no standardized way to characterise them because of their unique and complicated biochemistry. They have a three-dimensional conformation and surface-charge distribution which is dictated by the amino acid sequence (Kujundzic *et al.,* 2011). The charged surface and amino acids play a major role in determining the hydrophobicity of protein.

Fouling by protein can either be by adsorption or deposition. It is believed that adsorption fouling can both be in the internal or surface of the membrane. This takes place after the deposition of protein has occurred. Deposition on the membrane is usually on top of an already existing layer on the membrane which was formed by adsorption attachment to the membrane (Zeman and Zydney, 1996). A study by Mochizuki and Zydney (1993) revealed that the deposited layer of protein foulant on a membrane surface is compressible under high pressure operation. The compressibility of the protein fouling layer results in a decline in permeates flux due to the hydraulically resistant cake layer. The resistance of the protein layer in MF hydraulic demands for permeation are similar to that of UF due to increased packing density and compact fouling layer under high pressure operation. According to a study by Jonsson *et al*, (2006) a protein layer of 1 µm increases the fouling by a factor of 10 pH plays a major role in the extent to which a protein will foul a membrane. According to Zhang *et al.* (2002) the maximum adsorption of proteins onto the membrane occurs when the pH is at the isoelectric point. At the isoelectric point there is no net charge. When the pH is above the isoelectric point, the protein is negatively charged and if the pH is below the isoelectric, point the protein is positively charged (Velasco *et al.*, 2006).

Different concentration levels of protein in a solution have a different fouling mechanism on a membrane. Low concentration results in the internal fouling of the membrane while high concentration results in surface fouling of the membrane. Protein in high concentrations tends to aggregate forming larger aggregates that coat the membrane surface (Tracey and Davis, 1994). A study by Kelly *et al.* (1993) and many other researchers reported that increasing the concentration of protein in a solution results in a faster and stronger fouling layer being formed on the membrane surface.

(iv) Biofouling

Biofouling in simple terms can be defined as biological fouling. It is a net result of microbial attachment to the membrane and the subsequent growth and release of biopolymers that are associated with this microbial activity (Kujundzic *et al.*, 2011). Biofouling is also the most difficult to control as there is a large range of biofoulants that could be present in a particular aqueous system at a particular time for a particular feed solution. The foulants include proteins, organics, organic acids, polysaccharide fats, etc. Biofoulants in this section will be divided into humic materials and micro-organisms (bacteria).

Biofouling is the involvement of fine extracellular polymer fibrils that attach bacteria to surfaces (Marshall, 1976). The attached communities of bacteria in aquatic systems are encased in a glycocalyx matrix that is polysaccharide in nature, and this matrix material mediate adhesion (Coulter *et al,.* 1978). The biofilm is made up of single cells and micro colonies that are embedded in a highly hydrated, predominantly anionic exopolymer matrix (Jacques and Costerton, 1987). The attachment of bacteria to surfaces is irreversible and it results from a secretion by the bacteria itself which is a matrix of extracellular polymeric substances in which the bacteria cell are embedded in (Nguyen *et al.,* 2012).

The attachment of microbial cells to the membrane surface is the beginning of membrane biofouling. Subsequent to attachment of microbial, a biofilm layer is formed, which has a composition that is vast in diversity of different microorganisms which could be of bacteria, algae, protozoa, fungi, etc. (Flemming and Schaule, 1988). The sequence in which the biofilm is formed is first by adsorption of organic species and suspended particles on the wetted membrane surface to form a condition film. Thereafter, microbial cells are transported to the film. They attach to the membrane surface and the growth and metabolism of the attached microorganism and biofilm development (Al-Ahmad *et al.,* 2000). Extracellular polymeric substances (EPS) are high molecular weight secretions of microorganisms that consist of organic substances such as polysaccharide, protein, nucleic acids and lipids. EPS offer a binding base for biofilm to the membrane surface. They contribute to the mechanical stability of the biofilm and to the organisation of the biofilm community. Once the biofouling has been established other organic and inorganic materials contribute to the building of the fouling mass (Komlenic, 2010). EPS promotes the adhesion of microbial to surfaces by altering the physicochemical characteristics of the colonized membrane surface such as its charge, hydrophobicity and roughness. The EPS offers building blocks between the membrane pore and microbial cells. High concentration of EPS contributes greater binding capacity. Because EPS aggregates have many charged groups, they have both wetting and cross-linking characteristics containing both hydrophobic and hydrophilic sites on their structure, which enables them to be able to adhere to both hydrophilic and hydrophobic surfaces. The factor that makes biofouling very complex to understand and plan against is the fact that EPS have flexibility and rearrangement characteristics. This means EPS structure will rearrange themselves so that they are able to stick to any surface (Nguyen *et al.*, 2012).

Delgrange *et al.* (1998) found that biofouling from surface water was responsible for the 50% reduction of the membrane’s permeability in static conditions. The reduction was found to be more severe on hydrophobic membranes than hydrophilic membranes and pH had no effect at all. Deposition of a biofilm layer on the membrane surface was observed to affect more of the membranes with bigger pores than those with smaller pores. Though the reduction in flux was more on the hydrophobic membranes, the attachment of biofilm was not affected by the hydrophobicity.

2.8.3 Fouling mechanisms

Fouling of a membrane will arguably occur in two ways, namely macro solute adsorption (intermolecular interaction) and particle deposition (particle induced macro solute). These can take place progressively or simultaneously depending on the feed composition and membrane chemistry. It is the intermolecular forces between the membrane and solutes that governs membrane-solute interactions. These intermolecular interactions between the membrane and various components in the feed could be classed as quantum mechanical, electrostatic and polarization forces (Zeman and Zydney 1996).

Fouling roadmap for immersed membranes

The fouling roadmap for both constant pressure and constant flux operated systems take place in three progressive stages. In stage one is the constant pressure systems in which there is a rapid development of cake layer and membrane compaction. According to Field *et al.* (1995) this rapid cake formation on the membrane at the initial stages of filtration is due to the fact that the TMP required at the starting and maintaining reasonable flux is high. Flux at this stage is higher than the critical flux of the system, therefore the deposition rate of foulants on the membrane at this stage of filtration is high.

The first stage of fouling in constant flux systems is attributed to adsorption of SMP and EPS. Deposition in constant flux systems is not expected because most of these systems operate at subcritical flux conditions (Bacchin, 2004).

Following the cake development stage in constant pressure systems is the slow cake layer growth and further cake layer compaction. The low growth in the cake layer is explained by the theory of critical flux. The rate of deposition on the membrane during the slow cake growth stage is balanced out by back transport theory (Yoon *et al.*, 1999).

The second stage in constant flux systems is the slow rise in TMP. This is due to deposition of fine particles, SMP and EPS on the membrane surface. According to Foley *et al.* (1995) cross-flow velocity has little effect on fine particles. Therefore even though the system is operated below the critical flux, fine particles, SMP and EPS continue to deposit on the membrane resulting in pressure drop due to hydraulic resistance of the developed cake layer. Adsorption on a membrane is driven by the physical and chemical interaction between the foulants and the membrane. It in fact occurs at zero flux (Cho and Fane, 2002).

The final stage in membrane fouling is pseudo steady state for constant pressure systems and it is the sudden rise in TPM for constant flux systems. Both the final stages are explained by cake layer compaction. A study by Tarabara *et al.* (2004) explained that the compaction of the cake layer starts and exists from the bottom of the layer due to the squeezing force on the top layer to the layer beneath it. In constant pressure systems though at this stage the filtration process is at subcritical conditions and there is no further growth in the cake layer; however, the already developed layer causes the compaction of the bottom layer. This results in very small gradual decrease in flux. With constant flux systems the compaction of the cake layer is further compressed by the gradual rise in TMP in order to keep constant flux. The sudden jump in TMP is believed to be due to pressure drop across the membrane and cake layer reaching critical levels (Chang *et al.*, 2006). Membrane fouling takes place in four successive steps according to Belfort *et al.* (1992) as shown in Figure 2-16.

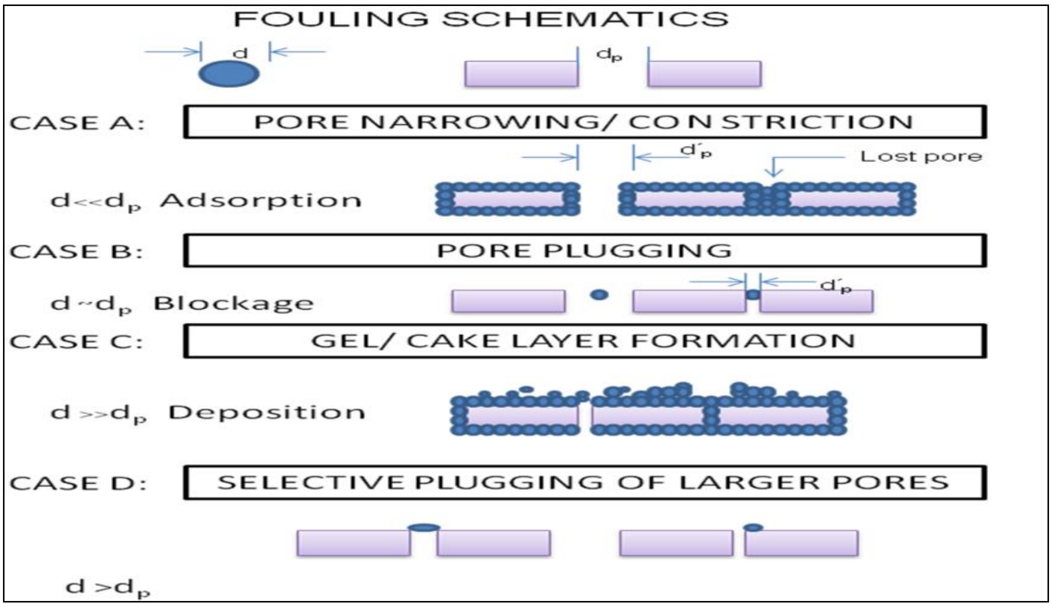

Figure 2-16 Fouling schematics by Belfort *et al.* (1992)

Case A: Particles plug smaller pores and reduce larger ones. This is usually as a result of adsorption particularly where there are fractions of protein in the feed solution.

Case B: Particles plug narrow pores. This can occur even in the absence of adsorption. Pore plugging normally occurs when a smaller particle enters a pore and along the length of the pore the particle gets stuck. Case B and Case A results in flow redistribution to other pores.

Case C: Particles form a layer on the membrane surface. This is as a result of deposition of material that supersedes the porous structure of the membrane. The membrane then becomes analogous to a pre-coat filter. Fouling is irreversible to the extent that the layer nearest the membrane is probably adsorbed onto it. Throughout the layer, the binding may or may not be irreversible.

Case D: Particles or debris plug the largest pores. This is observed in membranes with smaller pores than the filtering particles. The net effect here is that the larger pores get plugged leaving the smaller ones for filtration.

Pore narrowing results from the adsorption of protein like particles on the membrane internals, while pore plugging can also result from adsorption but is mostly due to colloidal material getting stuck on the pore channel as it travels with the permeate. Pore plugging by colloidal matter is favoured in membranes with bigger pores than the colloidal material. Protein or particle adsorption on the membrane is as a result of interactions between the membrane and the particles. Conditions that favour the interaction between the solutes and the membrane pore wall are when the fluid being filtered is a poor solvent and the membrane surface is highly energetic, like hydrophobic membranes in a protein containing solution (Belfort *et al.,* 1992).

Adsorption

Adsorption in membrane technology is the adhesion of solutes on both membrane surface and membrane internals. The attachment is facilitated by adsorptive forces between membrane and solutes. Factors influencing adsorption are the nature of the fouling particle (solute), solution properties and membrane material. Protein and natural organic matter (NOM) are the common foulants responsible for adsorption. Multi-component solutions are notorious for their fouling tendencies because they can contain both protein and NOM and many other foulants, thereby producing a very complex solution (Srebnik, 2003).

Dispersion interaction amongst the foulants and electrostatic repulsive forces between the foulants and the membrane governs the strength and the effects of adsorption fouling (Abdelrasoul *et al.,* 2013). Fouling by adsorption typically results in irreversible fouling and is amongst the first fouling mechanism on the membrane, because it starts fouling a membrane at zero filtration (Field *et al.*, 1995). Foulants that are notorious for adsorption fouling are protein like foulants and NOM. Once these foulants are attached on the membrane surface they tend to interact with each other due to Van de Waal forces. The Van de Waal forces pull the particles together and they aggregate into bigger particles on the membrane surface (Abdelrasoul *et al.* 2013). These then form a coat of aggregates on the membrane which blocks and reduces membrane pores resulting in a hydraulically difficult membrane to permeate through. As filtration progresses, more foulants are pulled by the Van de Waal forces onto the already attached foulants on the membrane surface and the membrane resistance increases. The electrostatic repulsive forces between the membrane and the foulants hinder the aggregation process of particles on the membrane. When the repulsive force is strong enough, the formed layer is porous and is loosely attached to the membrane. Consequently, the membrane permeability is not greatly reduced. However, the repulsive forces are generally not as strong as the dispersion interaction of particles on the membrane surfaces. This is why attachment of particles is always occurring in membrane filtration (Srebnik, 2003).

Adsorption fouling is also influenced by polar interactions. The interactions are between the membrane and particles, and are highly affected by the hydrophobicity of the particles and the hydrophobicity of membrane material. Polar interactions can either be attractive or repulsive. In the case where the polar interactions are attractive the presence of electrostatic repulsive forces hinders the strength of the attractive forces. The hydrophobic surface of the membrane and the foulants increases the attractive force between the two surfaces. The attachment that results from such interaction is strong and is unaffected by the repulsive electrostatic force. This is because most of the aquatic contaminants are hydrophobic. Hydrophobic membranes have been associated with high fouling rates due to adsorption or particle attachment on the membrane surface (Abdelrasoul *et al.,* 2013). The hydrophobic materials as explained by Roesink *et al.* (1990) tend to cluster together and as a result of clustering interfacial free energy are lowered which results in the high fouling rate of the two surfaces. Table 2-5 shows the classification of adsorption fouling in membranes.

Table 2-5 Classification of adsorption (Kohyama, 2012)

Adsorption Type	**Fouling Behaviour**	**Mechanism**	**Order of magnitude**
Physical adsorption	Reversible	Van der Waals forces	↓ Increasing Adsorption Strength
		Hydrogen-bond/ Hydrophilic, Hydrophobic	
	Quasireversible		
Chemical adsorption		Electrostatic force	
	Irreversible	Others	

Particle deposition

Particle deposition on the membrane is not possible in the absence of Brownian forces. The hydrodynamic drag forces acting on the particles as they approach the membrane is repelled by the membrane's electrostatic repulsive force. When this happens equilibrium is reached between the drag force towards the membrane and the electrostatic repulsive force from the membrane, if the forces are equal in magnitude. When filtration/permeation velocity is increased, the hydrodynamic drag force increases and the electrostatic repulsive force become overpowered by the drag force experienced by the particles. This also happens when the particles are close to the surface of the membrane. Brownian forces always tend to cancel the equilibrium promoting deposition on the membrane surface (Kim and Zydney, 2003).Brownian forces is a random movement of particles in a suspension. This results from the fluctuating forces which are a consequence of collision of suspended particles with the fluid molecules. The smaller the size of the suspended particles, the stronger is the effects of this force.

The electrostatic repulsive force is inversely proportional to separation distance. The further away the particles are from the membrane the stronger is the effects of the electrostatic repulsive force and the drag force is at its weakest at this position. Yet closer to the membrane surface the drag force is at its strongest and the Brownian force is also at its strongest, while the electrostatic repulsive force is weakest (Ramon *et al.* 2013).

Cake structure on fouling

A cake layer on the membrane surface is a result of deposition of foulants on the membrane surface. The permeability and structure of the developed layer is determined by the particle type, shape, size and interaction with the membrane material. Ramon *et al.* (2013) found that flat particles deposition rate on a permeable surface is much greater than spherical particles. This is because the repulsive force on flat particles is not as strong as it is on the spherical particles. The packing of flat particles is orderly and it results in a compact cake layer that is more resistant to permeation.

The permeability of a cake layer formed by fine particles of any shape is low due to its high packing density. The hydraulic demand for fluid flow through a densely packed cake layer is high (Foley *et al.,* 1995). The permeability of the cake layer decreases further under high permeating flux. This is because under these conditions the cake layer is being compressed by the high transmembrane pressure (Petsev *et al.,* 1993).

Smaller particles experience higher repulsive forces due to enhanced electrostatic double layer interaction, which makes the developed cake layer porous. The electrostatic double layer interaction is strongest with small particles. This is because small particles have fewer induced dipole moments (Kim and Hoek, 2002).

The cake layer that develops on the membrane surface under cross-flow mode is compact and highly resistant to filtration. This is due to the fact that cross-flow velocity is more effective in dispersing bigger particles. The fine particles are highly resistant to permeation and are left on the membrane surface and a cake layer develops with filtration time. As the cross-flow velocity is increased, the cut-off diameter of the particles that deposit on the membrane is reduced leading to even finer particles depositing on the membrane (Keskinler *et al.,* 2004).

Hwang *et al.* (2008) found that at low TMP operation fewer particles arrive on the membrane surface. If these particles are smaller than the pore size of the membrane, they enter the pores and block them. While under high TMP a large number of particles arrive simultaneously on the membrane due to the high flux. The simultaneous arrival of particles to the membrane surface hinders entry into the membrane pore and the net result is surface deposition of particles on the membrane surface.

According to Kuberkar and Davis (1999), cake layer on the membrane surface can be of benefit in mitigation against fouling by complex fluid/multi-component solution with high fouling propensity. The cake layer acts as the barrier between the membrane and the fouling components like protein, NOM and microorganisms. For example critical flux of BSA mixture is low, but when the mixture of BSA and yeast is used the critical flux of the solution is increased. This is because the yeast component, which has higher critical flux, coats the membrane and the BSA component gets attached to the yeast layer and not the membrane. The irreversibility of BSA fouling is reduced in the presence of yeast (Ye and Chen, 2005).

2.8.4 Concentration polarization

In the process of liquid passing through the membrane, particles accumulate in the vicinity of the membrane surface and form a thin layer. In the layer, the particles get stuck in the transverse direction with respect to the membrane surface. The stationary layer of retained particles provide additional resistance to the permeate flow. The resistance is dependent on the total number of particles in this layer and on the spatial distribution. The stationary layer containing retained particles is called concentration polarization (CP) and is inherent to all cross flow filtration processes (Song and Elimelech, 1995).

The retained solutes/particles diffuse back to the bulk solution. However, the rate of permeation in membrane systems is higher than the rate at which the rejected solutes diffuse back to the bulk solution. This results in a higher solute concentration at the membrane surface than in the bulk solution. As filtration progresses the concentration of the particles on the membrane surface becomes so high that a gel layer is formed which acts as a secondary barrier to permeate flux (Baker, 2004).

Concentration polarization is different for colloidal matter and for macromolecular solutions. CP for colloidal matter is minimal and has little effect on filtration because of a low value of osmotic pressure. The CP developed in the filtration of macromolecular solutions has a high value of osmotic pressure, as a result, reduction in filtration rate is observed. CP is a direct product of permeation or separation in membrane filtration. It is reversible and self-regulating. The self-regulating aspect is observed by the decline in permeate flux as the concentration polarization layer grows (Bacchin *et al.*, 2002). Figure 2-17 shows the formation of CP and fouling on membrane surface.

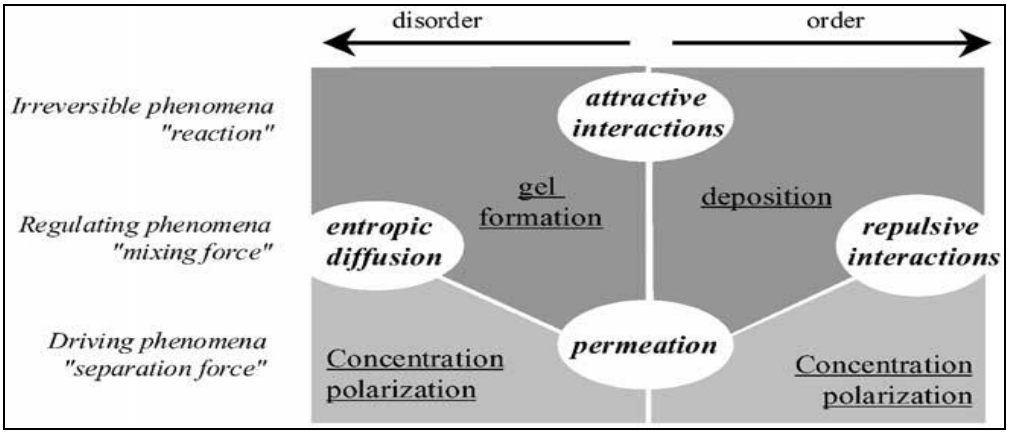

Figure 2-17 Formation of CP and fouling on membrane surface (Bacchin *et al.,* 2002)

In the filtration of macromolecular solutions according to Bacchin *et al.* (2002), concentration polarization is maintained by entropic diffusion. The entropic diffusion creates a disordered solid phase under totally attractive interactions. The disordered and permeable gel layer on the membrane surface is a result of the loosely deposited particulates on the membrane. Brownian diffusion forces between the particles in the solution, electrostatic repulsive forces from the membrane to the particles and the presence of bigger particles in a solution are the main contributors to the disordered gel layer formation, because the particles experience forces in all direction. However, if the driving force (TMP) and solute concentration is increased, the gel layer grows and its resistance to filtration increases leading to irreversible fouling and strong attachment of the gel layer to the membrane surface. This is because the developed layer under high TMP operation and high concentration is highly polarized, compact and orderly. Electrostatic repulsive forces acting on bigger particles are much greater and have a low packing density. This is the reason why with bigger particles the formed layer is porous and disordered (Bacchin *et al.,* 2002). Figure 2-18 show the CP of a macromolecule and colloidal material.

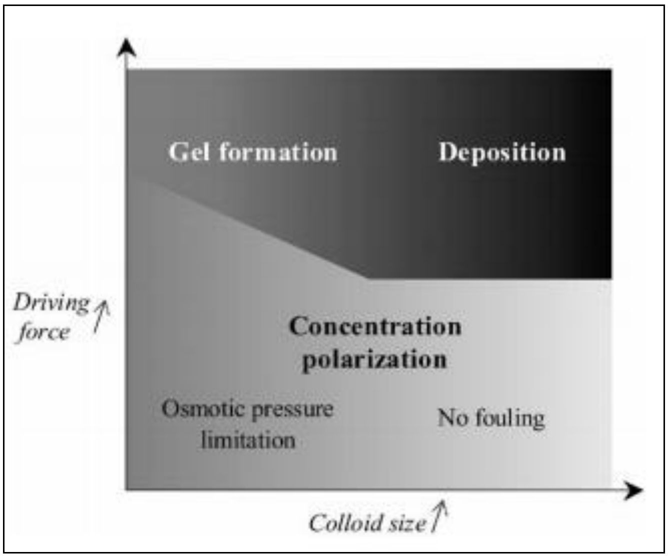

Figure 2-18 CP of a macromolecule and colloidal material (Bacchin et al., 2002)

Franken (2009) describes concentration polarization according to (equation 2-14)

$$\frac{C_m}{C_b} = \frac{e^{\left(\frac{J_V}{k_m}\right)}}{R + (1 - R).e^{\left(\frac{J_V}{k_m}\right)}} \qquad (2\text{-}14)$$

Where C_m is the concentration of the retained solute at the membrane surface (Kg/m^3), C_b is the bulk concentration of the solute (Kg/m^3), J_V is the volumetric flux through the membrane (m/s), k_m is the mass transfer coefficient for the solute (m/s) and R is the retention (in fraction) of the solute (Scott, 1995).

As seen in equation 2-14 according to Franken (2009) concentration polarization depend on a number of factors which are:

- **Filtration flux:** high filtration leads to concentration polarization because of the drag forces that forces the solutes to the membrane pores and surface. This leads to cake development and/or pore blocking, this phenomenon is controlled typically by cross flow velocity.
- **Mass transfer coefficient near the membrane surface:** high concentration of solutes at the membrane surface increases the effects of concentration

polarization. The most common way to increase mass transfer on the membrane surface is by cross-flow velocity. The movement of fluid adjacent to the membrane surface increases mixing at this point, reducing concentration polarization and improving mass transfer coefficient.

- **Retention of solutes:** high retention of solutes result in more pronounced effects of concentration polarization.
- **Bulk solution:** high bulk concentration of the solutes result in higher effects of concentration polarization at the membrane surface.

High driving forces give rise to irreversible fouling. This is true for both macromolecular solutes and also for colloidal particles whilst colloidal size and interaction with membrane surface plays a role in the way the deposition is formed on the membrane surface (Bacchin *et al.*, 2002).

2.9 Membrane fouling mitigation

Membrane fouling mitigation/control mainly deals with the management or minimization of the effect of membrane fouling since fouling itself cannot be completely avoided in membrane filtration (Franken, 2009). Membrane fouling can be controlled and managed at different stages. These are feed pre-treatment to limit its fouling propensity, improving the antifouling properties of the membrane, membrane cleaning and backwashing conditions and optimisation of operating conditions (Abdelrasoul *et al.*, 2013). It is common that the feed characteristics and the membrane properties cannot be altered to mitigate fouling, leaving only operation conditions for fouling control and mitigations. Figure 2-19 shows a decision tree or the most common way used in reducing the effect of fouling caused by the high concentration of the foulants in the feed.

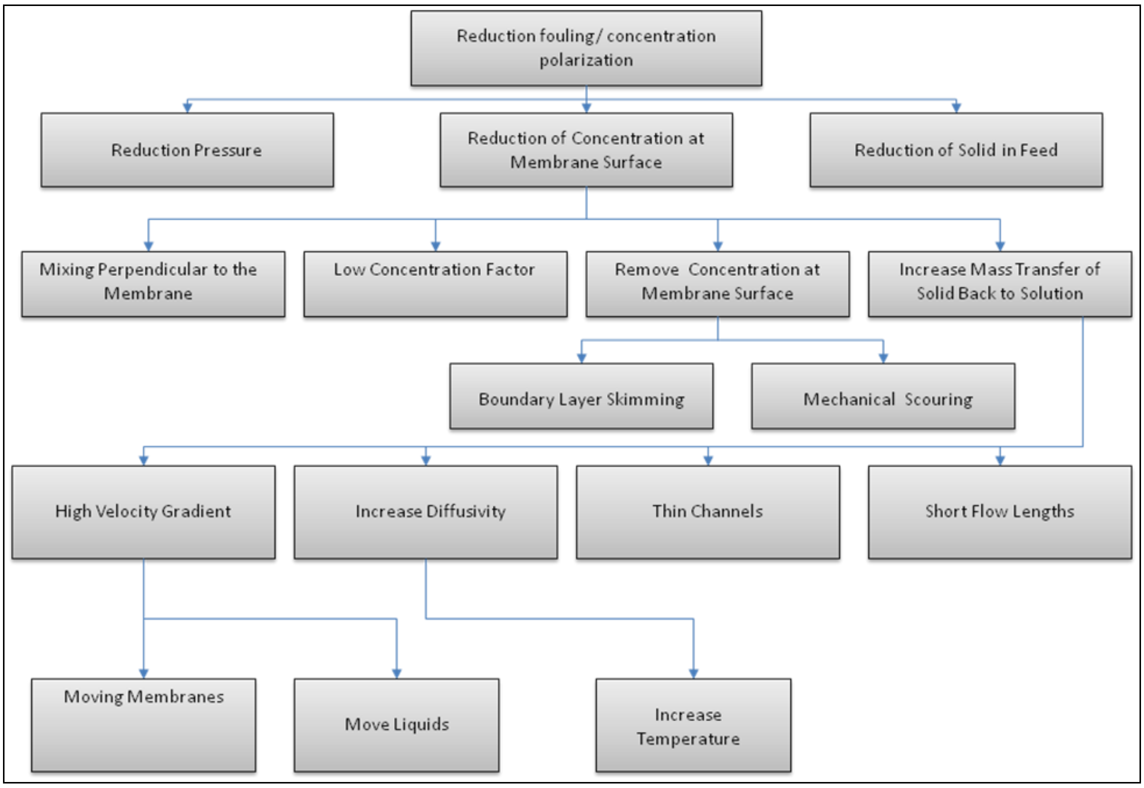

Figure 2-19 Approach to reduce the effects of CP and fouling (Bruin et al., 1980)

2.9.1 Temperature

Temperature of the solution is one of the parameters that can be altered to reduce the effects of fouling. However, this parameter is not often used for fouling control particularly in water treatment. Salahi *et al.* (2010) found that when the temperature of the oily wastewater was increased by 20 $^{\circ}$C there was an increase in flux of about 60%. This was attributed to an increase in the diffusion rate as the temperature was raised.

2.9.2 Cross-flow velocity

Cross-flow velocity is the most common and widely used method to control fouling at the membrane surface. The method works by pumping the feed stream at a superficial velocity parallel to the membrane surface (Abdelrasoul *et al.*, 2013). High cross-flow velocity creates mixing on the membrane surface thereby improving the mass transfer coefficient, but the increase in mass transfer coefficient is different for different feed solutions (Scott, 1995). At the membrane surface for any filtration system, rejected particles accumulate in a boundary layer. According to Ficks' law, particles in the boundary layer collide with each other more frequently improving particle diffusion from the boundary layer to the bulk solution. This diffusion can be improved by what is called shear induced diffusion. This is achieved by causing movement of the liquid close to the boundary layer. When the movement of the liquid is increased, the particle collision becomes vigorous and the particle diffusivity is increased (Eckstein *et al.*, 1974). Shear induced diffusion of particles is highest at the membrane surface or at the boundary layer because of the high particle density in that region (Zydney and Colton, 1986).

Cross-flow filtration systems can be operated in either laminar, transition or turbulent region. A study by Cui *et al.* (2013) showed that the best results are obtained when the system is operated in the transition cross flow velocity. This was found to be due to the fact that in this region there is no stagnant layer that develops on the membrane surface and shear induced diffusion is maximized. Whereas when the system was operated in the laminar region a stagnant layer was allowed to develop on the membrane surface due to the laminar flow. And turbulent region is not constant through the length of the membrane. As a result, one end of the membrane performs well but further away from the pump/membrane entry, the membrane is operated in a laminar region. According to Singh (2007), increasing cross-flow velocity improves flux provided the transmembrane pressure is not increased to the same magnitude as the cross flow velocity.

The pumping requirement for cross flow systems to control fouling is energy intensive. This has contributed to the limited growth of membrane technology (Abdelrasoul *et al.*, 2013). Besides the energy requirements for the traditional cross-flow systems, many membrane researchers have found that the intensive shear stress offered by cross flow systems through pumping devices, breaks the suspended particles/microbial flocs. The resulting fine particles are highly fouling, and in the case of wastewater treatment breaking of microbial flocs releases EPS resulting in catastrophic fouling occurring on the membrane (Kim *et al.,* 2001).

2.9.3 Aeration

An alternative to cross flow filtration is the immersed membrane systems. In this system, vacuum pressure is applied to get permeate while from the bottom of the membrane pack air is supplied to scour the membrane surface and inhibit deposition on the membrane surface (Yamamoto *et al.,* 1989). As with cross-flow velocity, air sparging/scouring induces shear force at the membrane surface as the air bubbles rise travelling adjacent to the membrane surface. The mechanisms responsible for the shear force in the membrane surface are fall film effect and wake effect. These are a net result of the quick rise of air bubbles and the feed solution (Judd *et a.,* 2001; Berube *et al.,* 2006; Klegraf and Lahnsteiner, 2008).

Air scouring generates localized cross-flow conditions along the membrane surface thereby reducing the deposition of particles and the development of a cake layer on the membrane surface. The same effect of shear induced diffusivity of the particles seen in cross flow velocity is achieved in immersed membrane systems but with air bubbles as the media to promote particle collision on the membrane surface. In immersed membrane systems when the air flow rate is increased, shear intensity on the membrane surface is increased and the fouling effects are reduced (Delgado *et al.,* 2008). Also increasing the aeration rate has a critical high limit, above which no fouling reduction is observed. The suspended solids and microbial flocs above the critical aeration rate break. When this happens, microbial flocs release EPS and the suspended solids are broken into fine high fouling particulates (Ivanovic and Leiknes, 2008).

2.9.4 Relaxing, back flushing/ backwashing

Intermittent relaxation of the membrane during filtration helps loosen the fouling layer on the membrane surface and makes the deposited layer on the membrane surface permeable. The duration and the frequency of the relaxation play a role in fouling reduction. Long relaxation periods result in high instantaneous fluxes which result in high fouling rates and long run periods before relaxing. The system interprets them as continuous dead end operation and lead to a compact layer on the membrane surface (Wu *et al.,* 2008).

Back-flushing involves pumping permeate water at a high cross-flow velocity in the opposite direction from which the feed comes in. The high velocity used for back flushing, is supposed to release and discharge the adsorbed fouling on the membrane surface. This method is effective in removing colloidal particles trapped on the membrane surface (Arnal *et al.*, 2012). Back-flushing of the membrane plays the role of loosening the cake layer on the membrane surface as well as unblocking of the membrane pores. A study by Cakl *et al.* (2000) showed that the biggest benefits of back flushing are seen at shorter back pulse, high transmembrane pressure and lower cross flow velocity.

Backwashing is a reversed filtration process in which the permeate of backwashing solution is flushed through the membrane back to the concentrate side. The pressure in the permeate side is made higher than the pressure in the membrane pores and concentrate side. This allows membrane pores to be cleaned (Arnal *et al.,* 2012). A study by Delgado *et al.* (2008) showed that backwashing is moderately successful in removing internal clogging material from the membrane internals. When backwashing was coupled with surface flushing even better results for backwashing were observed. It was seen in their study that the duration of a backwash is more effective in limiting the effects of fouling than the frequency.

2.9.5 Transmembrane pressure

Transmembrane pressure controls the transportation of particles to the membrane surface. Cross-flow velocity or air scouring promotes the back transportation of particles from the membrane surface to the bulk solution thereby improving mass transfer at the membrane surface. High transmembrane pressure results in high permeate flux which leads to quicker deposition on the membrane because the membrane system is being operated above critical flux (Abdelrasoul *et al.,* 2013). Besides facilitating the transportation of particles to the membrane surface, high TMP also reduces the permeability of the already deposited fouling layer on the membrane surface. This further increases the effect of membrane fouling (De Bruijn *et al.* 2005; Chang *et al*,. 2006).

To avoid excessive fouling, if the TMP is increased the cross-flow velocity must be increased in order to keep the system below critical flux (Bacchin *et al.* 2002). However, Huang *et al.* (2012) found that high transmembrane pressure in microfiltration flushes the concentration polarization layer through with the membrane pores to the permeate side. Hwang *et al.* (2008) found that high instantaneous flux causes large amount of particles to simultaneously arrive on the membrane surface. The particles arrive at the same time, entry to the pore internals of the membrane is hindered. This results in particles depositing on the surface of the membrane rather than the pores. Surface membrane fouling is reversible while internal fouling is mostly irreversible and is destructive.

2.9.6 Membrane surface

Various studies have shown that interactions between the membrane and the foulants in the feed solution play a major role in membrane fouling. Lee *et al.* (2004) showed in their study that hydrophobic membranes are more prone to fouling by aquatic pollutants found in surface water. Membrane pore size also affects fouling. According to Hwang *et al.* (2008), membranes with big pore size are more susceptible to internal pore plugging than those with small pore sizes. However, Bacchin *et al.* (2002) found that the effects of concentration polarization and gel layer fouling is more pronounced in membranes with small pores like UF, NF and RO and not so much on MF.

The effect of membrane surface roughness as studied by Hashino *et al.* (2011) showed that rough membrane surfaces are more prone to fouling than smooth surface membranes. This was attributed to greater surface area available for attachment by the foulants and stagnant areas which are inaccessible by cross flow velocities and/or air scouring.

Modification of the membrane surface is usually done at the manufacturing stage of the membrane. This is done by impregnating the membrane with fouling retarders. At the application stage Satyawali and Balakrishnan (2009) coated the membrane with powdered activated carbon and the critical flux of the system was increased. Mecha and Pillay (2014) also impregnated a highly hydrophobic microfilter with nano-silver. The hydrophobicity of the membrane was reduced and the membrane pure water flux increased.

2.9.7 Cleaning

Membrane cleaning is an integral and an important part of membrane processes. Cleaning could either be done hydraulically or chemically. Membrane cleaning becomes necessary when avoiding irreversible fouling of the membrane.

Hydraulic cleaning

Hydraulic cleaning uses mechanical forces to dislodge and remove foulants from the membrane surface (Arnal *et al.,* 2012). The most common methods to hydraulically clean membranes are through backwashing and sponge ball cleaning. These methods of cleaning are typically used in cleaning in place (CIP) situations. Backwashing has already been discussed above. Sponge cleaning is done mostly in tubular membranes where the module diameter is big enough for this type of cleaning. A sponge is inserted into the membrane module to scrub off the foulants from the membrane surface (Arnal *et al.* 2012).

To enhance backwashing, membrane cleaning chemicals can be added to the permeate/solution to be used for backwashing (Abdelrasoul *et al.,* 2013). According to Holman and Ohlinger (2007), backwashing is less effective in dealing with adsorption fouling. However, if oxidizing chemicals or disinfectants are added to the backwashing solution, flux recovery is usually better.

Chemical cleaning

When membrane fouling can no longer be completely removed by physical cleaning, the membrane is irreversibly fouled. Such fouling can only be chemically removed. Caution is to be employed when chemically cleaning the membranes because the membranes also can be damaged by the chemicals used for membrane cleaning (Gao *et al.,* 2011). The choice of chemicals for membrane cleaning must be able to completely dissolve the foulants on the membrane but not damage the membrane itself (Arnal *et al.,* 2012).

Chemical cleaning is a reaction between the chemicals and the foulants on the membrane surface. The process involves mass transfer of the chemicals to the fouling layer and the products of the reaction are dispensed back to the bulk liquid phase. Effectiveness of the chemical cleaning is improved by hydrodynamic conditions that promote contact between the cleaning chemicals and the fouling layer on the membrane surface. This is mostly done by circulating the cleaning solution over the fouling layer (Abdelrasoul *et al.*, 2013). Table 2-6 shows the fouling constituents that impacts fouling and their mechanisms on the membrane. Table 2-7 show the types and function of the cleaning agents. Table 2-8 give examples of foulants and fouling modes in membranes.

Table 2-6 Fouling constituents that impacts fouling (Abdelrasoul *et al*., 2013)

Fouling Category	**Responsible constituents**	**Fouling Mechanism**
Inorganic/ Scaling	Metal Oxides, Inorganic colloids, Calcium Sulphate, Carbonate, Fluoride, Barium Sulphate, Silica	Pore narrowing, Gel/Cake formation
Particulate Fouling	Suspended Solids, Colloids, Biologically Inert Particles	Pore narrowing, Pore plugging
Biological/Microbial Fouling	Bacteria, Microorganisms, Concentration Polarization	Pore narrowing, Pore plugging, Gel/Cake Formation-most prominent
Organic Fouling	NOM	Pore narrowing, Gel/Cake formation

Table 2-7 Cleaning agents (Holman *et al.*, 2007)

Chemical cleaning category	**Major Function**	**Typical Chemicals**
Caustic	Hydrolysis, Solubilisation	NaOH
Oxidants/ Disinfectants	Oxidation, Disinfection	NaOCl, H_2O_2, Peroxyacetic Acid
Acids	Solubilisation	Citric nitric, Hydrochloric acid
Chelating Agents	Chelation	Citric acid, EDTA
Surfactants	Emulsifying, Dispersion, Surface conditioning	Surfactants, Detergents

Table 2-8 Examples of foulants and fouling modes in membranes (Scott, 1995)

Foulants	**Fouling mode**	**Fouling mechanism**
Suspended particles	These particles come in with the feed and are developed on the membrane surface due to particle to particle interaction	Membrane channel/ pore blockage as well as cake layer formation
Colloidal particles	Come in with the feed	Pore internal blocking, slimy layer development (gel layer)
Macromolecules	Gel or cake formation on membrane.	Macromolecular fouling within the structure of porous membranes
Small molecules	Some small organic molecules tend to have strong interactions with some polymeric membranes	Adsorption fouling/ adhesion on membrane material
Proteins	Interactions with surface or pores of membranes	Adsorption fouling/ adhesion on membrane material
Chemical reactions leading to scaling	Concentration increase and pH changes can lead to precipitation of salts and hydroxides	Membrane structural change, scaling
Biological	Growth of bacteria on the membrane surface and excretion of extracellular polymers	Adsorption fouling/ adhesion on membrane material

2.10 Summary

The main purpose of treating drinking water is to bring it to an acceptable quality level for human consumption, by removing microorganism (bacteria) and viruses that could cause harm to human health. The removal of these dangerous microbes in the conventional water treatment process is achieved mostly in the filtration and disinfection processes. Conventional water treatment facilities are a combination of different treatment processes commonly arranged in a train sequence. However there are fundamental limitations of conventional water treatment facilities to meet current water quality requirements because of the ever changing quality of the feed, the centralize nature of the conventional water treatment facilities poses other challenges all together.

The superiorities of membrane technology over the conventional water treatment plants include but are not limited to; flexibility to upgrades and scaling, ability to handle varying incoming quality, small foot print plants, can be decentralized and centralized at will, can be easily incorporated into existing water treatment facilities and produce superior quality than most of the competing technologies. The advantages offered by membrane technology have made it suitable for rural communities where topographies are difficult and there is no infrastructure that conventional water treatment plants require. Lack of accurate information on the quality of the raw water to be used with the membrane at design stages has enhanced the risk of membrane fouling in drinking water treatment. Membrane fouling is a major concern in drinking water treatment systems because high cost and membrane integrity issues are incurred whilst trying to maintain the membrane. Commercially available membranes are very sensitive to rough treatment resulting in frequent cycles and maintenance costs. Membrane fouling contributes to 80% of the operational and maintenance costs of membrane systems; this is even higher than the skills of the personnel operating membrane systems. Operation and maintenance cost of membrane systems could be higher than the capital costs of the system.

Membrane fouling mitigation/control mainly deals with the management or minimization of the effect of membrane fouling since fouling itself cannot be completely avoided in membrane filtration. Current available membranes are not suitable for rough treatments and as a result the user needs to keep strictly to the maintenance procedures supplied by the manufacturers if the full benefits of the technology are to be enjoyed. This was identified as a gap in the technology by the Department of Chemical Engineering, Membrane Water Research Group at Durban University of Technology. This then led to the evaluation of a woven fibre micro filter membrane (WFMF). The membrane is made of a polyester based fibre material, locally supplied by the Gelvenor Consolidated Fabrics (PTY) Ltd in Durban, South Africa. The membrane has high mechanical and tensile strength, it is robust and inexpensive and readily available locally. This membrane is suitable for under developed countries where the technical know how to operate and handle complex and sensitive systems is limited. The separation integrity of the material was found to be satisfactory for the purpose of drinking water treatment.

This study is focused on evaluating the performance of the woven fibre membrane filtration unit with respect to its fouling propensity to different feed samples. It is also to evaluate and developed flux enhancement and cleaning strategies and flux restoration after fouling.

Chapter-3 METHODOLOGY AND METHODS

3.1 Introduction

This section describes briefly the equipment and procedures that were used. Experimental work was conducted at the Durban University of Technology (DUT) laboratory, using a synthetic feed. Raw water synthetic feed was used because it was not convenient to bring large quantities of Duzi/Umkomaasi River samples to the DUT laboratory due to logistical problems. These rivers were situated out of the Durban metropolis. The criterion for the materials, study approach, analysis and experimentation and methods are described in detail.

3.2 Materials

The major materials used for this study was the feed samples and the membrane materials. These are described briefly in the following sections.

3.2.1 Synthetic feed

The synthetic feed was made to be of similar fouling characteristics with the local rivers Duzi and Umkomaasi Rivers in Durban, KwaZulu-Natal. The reason for choosing these specific rivers was their fouling abilities on the WFMF membrane. The two rivers were analysed to obtain the parameters responsible for WFMF fouling. Similar parameters were then used in similar quantities, to the two rivers, to prepare the synthetic feed.

The synthetic feed solution used in this study was made up of river clay, domestic sewerage, and tap water. The concentration of river clay in the synthetic feed was 3 g/ℓ and the domestic sewerage was maintained at 2% of the total volume of the synthetic feed solution prepared. The basic composition of the feed solution is presented in Table 3-1. The size of the river clay particles were measured to be 0.4 ± 0.05 microns using a modified size exclusion method (MSE). The final composition of the synthetic feed solution was arrived at using a trial and error method, so that its fouling profile on a clean membrane was similar to that of Duzi and Umkomaasi Rivers. The identified major foulants from Duzi and Umkomaasi River were colloidal matter and microbiological components. This was the reason clay and domestic sewerage was used in preparing the synthetic feed. The residual chlorine of the tap water in the feed tank was measured once a day and was maintained below 0.1 mg/ℓ. This was to ensure that microbiological deactivation does not occur in the feed tank.

Table 3-1 Characteristics of feed samples

Contaminants	**Units**	**Duzi River**	**Umkomaasi River**	**Synthetic Feed**
Turbidity	NTU	80	120	200
Colour	^{0}H	8.5	9.5	8.5
Total coliforms	Count per 100 mL	19344	12300	15475
E.coli	Count per 100 mL	8412	2140	12324

3.2.2 Membrane

A flat sheet woven fibre microfiltration membrane (WFMF) was used for this study. The membrane is a polyester based fibre locally sourced and was supplied by the Gelvenor Consolidated Fabrics (PTY) Ltd in Durban, South Africa. The pore size of the membrane was 0.45 µm inferred from its similar separation abilities with a 0.45 µm millipore filter paper and a 0.45 micron Kubuta membrane.

Table 3–2 Equipment specifications

Symbol of equipment.	**Equipment Name**	**Equipment specifications**	**Operation range**
P1	Meter Centrifugal pump (CPM128/130)	*HP*:0.5kW:0.37, Q_{max}:35L/min, H_{max}:35m	Operated intermittently at 5 - 20 L/hr.
P2	Donken fans (500-1.0-5.5)	*HP*: 5.5, Air Q_{max}: 1980 L/hr., Maximum pressure: 5.7 kPa	Air Q_{max} =1.8 – 5.4 m^3/hr. at pressure of 4.5 kPa
FI1	Kytola adjustable air flow instrument	10- 150 L/min	30 – 90 L/min
V2	Brass throttle	1 inch throttle valve	
LC1/2	Float level controller	½ inch throttle valve	
Diffuser	PVC air spurger	1 inch PVC pipe with 5 mm holes	
Stirrer	RW28 IKA stirrer	100 L; 1400 rpm	300 rpm

3.2.1 MSE method

The modified size exclusion method (MSE) was used to determine the sizes of the clay hparticles. The test was done on the Sterifil Aseptic Vacuum System with a Hand vacuum. Three millipore filter papers of different pore diameters were used on the vacuum filter equipment to filter a dilute clay solution. The different filter paper's pore diameters were 1, 0.45 and 0.2 µm. The turbidity of the dilute clay solution was measured using HACH 2100Q Portable turbidi meter, before and after filtration. The filter paper that retains all of the turbidity was regarded as having a smaller pore diameter than the clay particles. The filer paper that allowed all of the turbidity to pass through was regarded to have a bigger pore diameter than the clay particles. The particle size of the clay was regarded to be in-between the filter paper that allows 50% and more of the turbidity and the filter paper that rejects 100% of the turbidity. Figure 3-1 shows the microscopic view of WFMF membrane at X 1000.

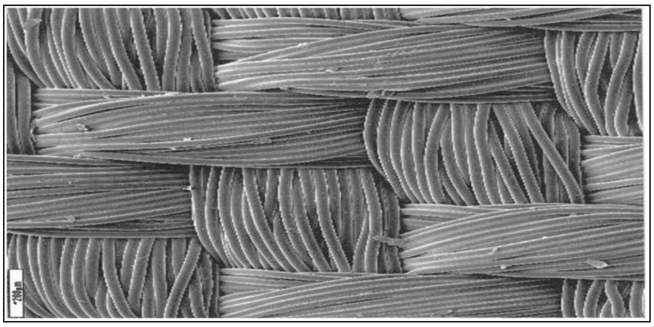

Figure 3-1 Microscopic view of WFMF membrane at X1000

3.2.2 Procedure for module making

A plate and frame module that was 500 mm in length and 300 mm in width was constructed on a 5 mm PVC frame. In between the two sheets of membrane materials a wire mesh was inserted to enhance the fluid flow inside the membrane (see Figure 3-2). The modules were packed together into a pack of 5 modules and spaced 5 mm apart from each other. The pack was held together by four carbon steel rods that were inserted through the holes drilled on the membrane edges and 5 mm nuts were used to separate these membrane modules. The total filtration area of the membranes was 1.5 m^2. Each membrane module had its individual permeate collection point outside the filtration tank. This was used for individual analysis and data verification. The thickness of the membrane material was 1.2 mm and was measured by a Vernier Calliper. A schematic of the membrane module is shown in Figure 3-2.

Figure 3-2 Photo of a membrane module

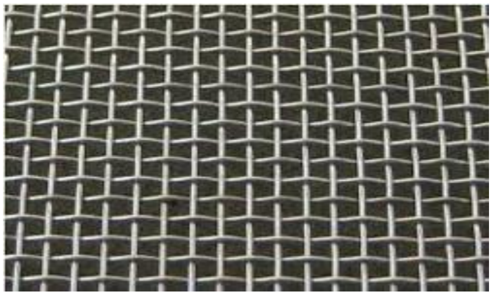

Figure 3-3 Membrane spacer

3.2.3 Analytical test

Turbidity tests were carried out with the HACH 2100Q Portable Turbidi meter. The standard method of using the turbidity meter was followed (HACH Portable Turbidimeter, 2008). The procedure to use HACH 2100Q Portable Turbid meter is attached in Appendix D1.

E. coli and coliform tests were done according to the IDEXX Collilert reagent method. The details of the method can be seen in the appendix D2.

Total and free chlorine was tested using HACH new 5870000 Pocket Colorimeter. The DPD method used was and the details of this method is presented in appendix D3

3.3 Experimental setup

Figure 3-4 shows the schematic flow diagram for the experimental setup as conducted. The membrane pack of five membranes was fully immersed into a 100 L filtration tank. The filtration was gravity driven and the head above the membranes was maintained at

0.4 m by a float level controller LC1. The filtration tank was fed by gravity from a 200 L feed tank via level controller LC1. The feed solution was kept constant by gently mixing the tank with a stirrer set at 300 rpm. Care was taken to ensure that vigorous mixing in the feed tank did not occur which would have resulted in particle breakage. The permeate was collected at the bottom of the filtration tank via a common manifold situated outside the filtration tank into a permeate tank. Some of the permeate from the permeate tank was pumped by a centrifugal pump, P2, to the backwashing tank to maintain the level when backwashing the membranes. The rest of the permeate from the permeate tank was allowed to flow to the drain.

The external membranes (Membranes 1 and 5) within the membrane pack were used as the base fouling case for each experiment, hence these two membranes were not exposed to any flux enhancement strategies. The permeate flow was manually measured via V2, where the time to fill a specific volume was timed. The filtration rate was adjusted on each individual membrane. The membrane scouring air was supplied by a centrifugal blower. The air was supplied from the bottom of the filtration tank via an air diffuser, which had 0.5 mm diameter holes. The air diffuser was positioned in-between the membranes so that the rising bubbles could make contact with the membrane. The air scoured membranes together with the air diffuser were boxed in together by baffles to prevent air from escaping and not making contact with the membranes. The scouring air flow rate was measured by flow-meter FI1. The air flow rate was adjusted manually via V2.

When the membranes were not being backwashed, the backwashing individual valves located at the top of each membrane were shut and the entire permeate was sent to the drain. The individual permeate valves at the bottom of each membrane module were to enable individual membrane isolation and to measure permeate flow measurement for each membrane individually. Individual backwashing valves LC2 were installed to enable backwashing of the membranes separately. LC2 was to ensure that the backwashing head pressure was maintained at a constant of 0.5 m.

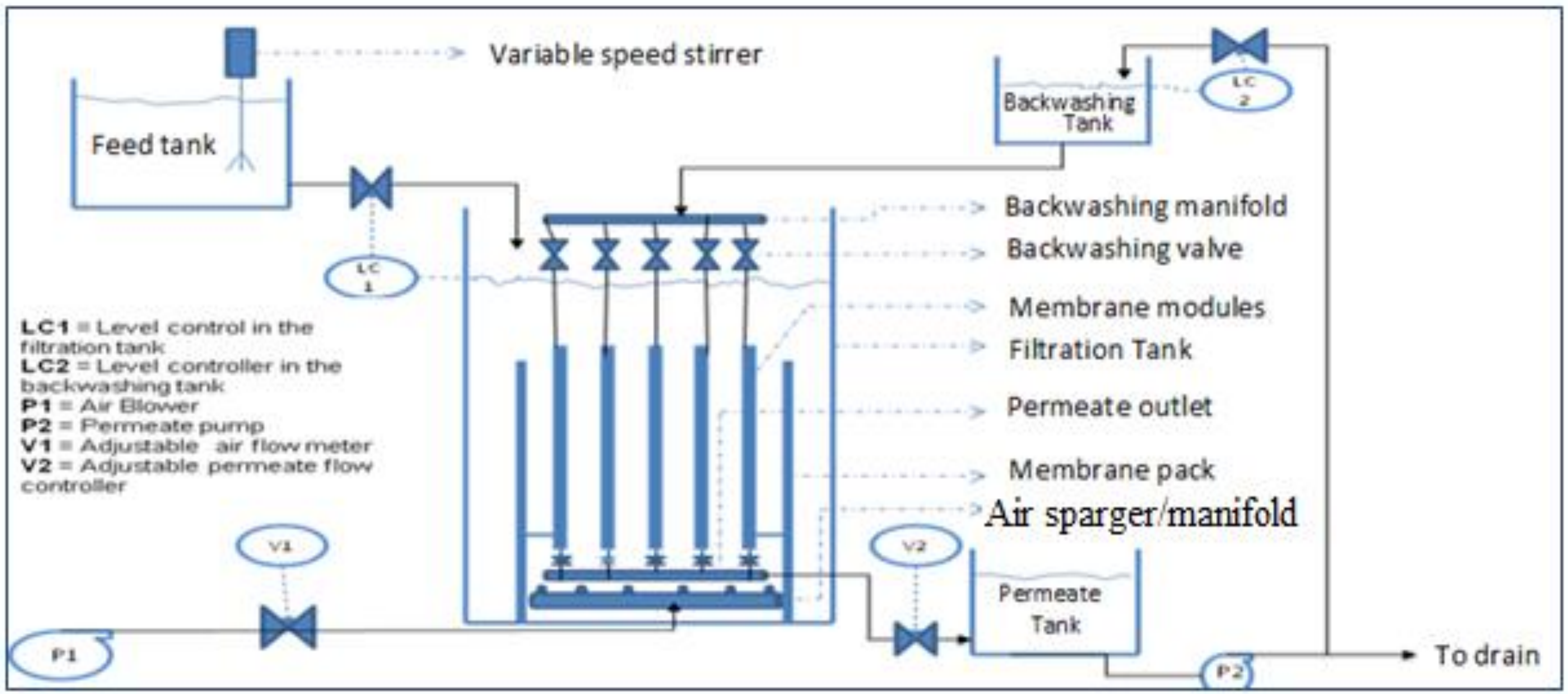

Figure 3-4 Experimental set-up

3.4 Experimentation procedure

The study was divided into four sections; (1) WFMF membrane and feed solution fouling characterization. (2) Assessment of various flux enhancement strategies on the WFMF membrane; (3) Optimization of the flux enhancement strategies (4) Evaluation of various cleaning strategies on WFMF membrane. A brief procedure on how each experiment was conducted is presented in section 3.4.1.

3.4.1 Procedure for WFMF membrane pore determination

The traditional methods to determine the pore size of a membrane, such as a bubble test and microscopic view were unsuccessful due to the configuration of the WFMF membrane (see Figure 3-1). Therefore, the pore size of the WFMF membrane was inferred to a filter paper that exhibited similar separation efficiency. 0.5 g/ℓ of $Ca(OH)_2$ was dissolved in de-ionized (0.1 ± 0.05 NTU) water in a 30 litre container.

The particle size of the $Ca(OH)_2$ was 0.8 micron. The $Ca(OH)_2$ solution was passed through three different millipore filter papers (1, 0.45 and 0.22 µm). The filtration pressure was kept constant at 10 kPa. The experiment set up is shown in Figure 3-5. The turbidity of the feed solution and that of the filtrate were measured and recorded. The same experiment was repeated but with the WFMF membrane in place of the filter paper. Feed and filtrate turbidity were measured and recorded. The filtrate turbidity of the WFMF membrane was compared to that of the individual filter papers and that of de-ionized water to check for percentage rejection. The pore size of the WFMF membrane was inferred to be equal to the filter paper with similar rejection capacity.

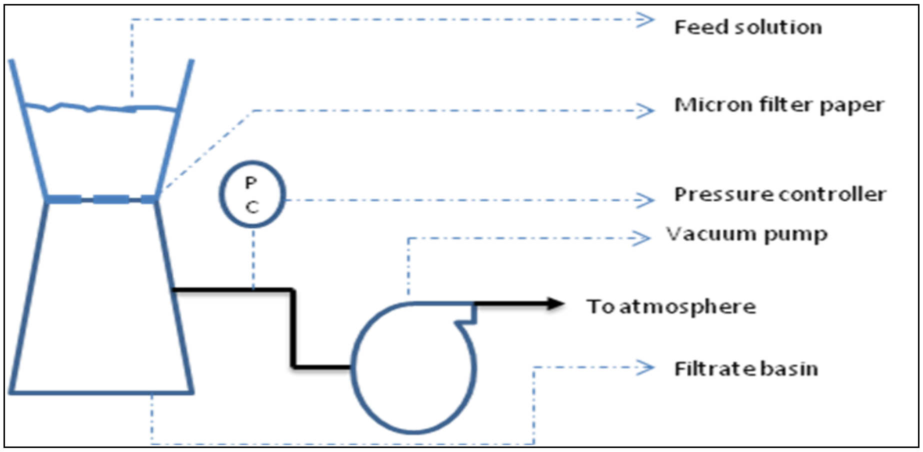

Figure 3-5 Filtration cell for 1 um filter paper

3.4.2 Adsorption test of WFMF membrane with the synthetic feed

Adsorption fouling occurs on the membrane even in the absence of filtration. To evaluate adsorption fouling on the WFMF membrane, a PWF test was carried out on the clean WFMF membranes and the obtained flux was recorded. Immediately after the pure water flux, the membranes were immersed into the synthetic feed solution. Care was taken that there was a minimum head above the WFMF membranes. This was to ensure that filtration does not occur. The WFMF membranes were immersed horizontally such that only 1 cm head was available to drive filtration. The membranes were immersed for 20 minutes after which the pure water flux was carried out again to see whether there was any loss of flux due to adsorption. Figure 3-5 shows the filtration cell used for the determination of adsorption fouling on the WFMF.

3.4.3 WFMF membrane and feed solution fouling characterization

Fouling characterization experiments were conducted to determine the flux decline rate under constant flux operation. Six different samples were filtered and the performance of the WFMF membrane with respect to flux decline was observed. The samples used were Duzi River, Umkomaasi River, Synthetic feed, clay solution, 0.8 µm $Ca(OH)_2$ solution and 2 µm Ca(OH)2 solution. The clay solution, 0.8 µm Ca(OH)2 solution and 2 µm Ca(OH)2 solution were at a concentration of 3.5 g/ℓ dissolved in tap water.

Duzi and Umkomaasi River were found to be highly fouling on the WFMF membrane. Therefore, these two rivers were used as a standard to prepare the synthetic feed. The major foulants that were common in these two rivers were turbidity (suspended colloidal matter) and microbiological components (coliforms and bacteria). Therefore, the synthetic feed was prepared with Duzi River clay and activated sludge from Northern Wastewater Treatment works, Durban, South Africa. The fouling behaviour for the 3 feeds was studied on a 40 L membrane filtration cell as shown in Figure 3-6.

Water from Duzi and Umkomaasi was poured into the membrane filtration cell. The filtration cell was filled to the top to ensure that the membrane was completely immersed. Flux measurements were taken every 10 minutes for a duration of 2 hours. The flux results were plotted on a flux time graph to observe the flux decline over time. The membrane filtration cell was gravity driven.

The experiment was repeated for the synthetic feed solution. A 50 L solution as prepared for each experiment. The prepared solution was filtered with the membrane filtration cell and flux against time profile graphs were plotted. The synthetic feed flux profile was compared to the flux profiles of Duzi and Umkomaasi River. The composition of the synthetic feed was adjusted until the flux profile was similar to the profiles of the two rivers. The activated sludge was kept in the refrigerator at 10°C for a maximum of 5 days. Thereafter, new sludge was brought in. This was to ensure that the biological fouling characteristics of the microorganisms were kept constant for the duration of the experiments. The filtration area of the WFMF membrane used was 0.05 m^2.

3.4.4 Fouling

Fouling measurements or indicators for the entire study was based on flux decline against time. The operating head pressure was maintained at 0.4 m. The decline in flux for a constant pressure membrane system is as a result of membrane fouling (Abdelrasoul *et al*, 2013). Flux measurements were taken throughout the day. The system was then left to run over night and the cycle was repeated the next morning. The profiles for flux enhancement strategies used were plotted on daily averages.

3.4.5 Fouling test

Fouling is said to have been established on the membrane, when the filtration driving force is removed and the original membrane flux is not restored (Bacchin *et al.,* 2002). To evaluate fouling on the WFMF membrane, the following steps were done. Normal feed filtration was stopped and the membranes were rinsed with tap water in order to remove all the loosely deposited particles on the membrane surface. Once the loose fouling on the membrane was removed PWF was evaluated. The PWF was carried out using water as the feed solution, the filtration was carried out using the rig system, shown in Figure 3-6. The difference between the pure water flux of a clean membrane pack and that of a fouled but rinsed membrane pack, gave an indication of the fouling on the membrane pack.

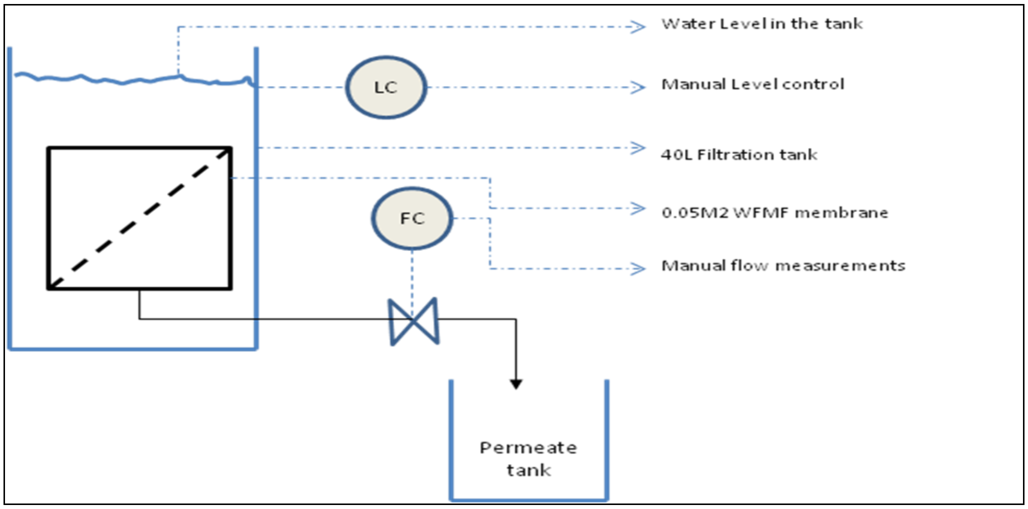

Figure 3-6 Schematic flow diagram for fouling test

3.4.6 Gradient to assess cleaning and flux enhancement strategies' performance

In this study, the gradient was used to assess the rate of flux decline. A high value of the gradient indicates the rapid decline in flux over filtration. The rapid decline in the permeate flux can also be interpreted as high fouling of the WFMF membrane with filtration time. According to Yoon (2011) flux decline in constant pressure driven systems occurs in three progressive steps; severe drop in flux, moderate drop in flux and very slow drop in flux. This is when no flux enhancement strategy is used. To study the effectiveness of each flux enhancement strategy, gradients were calculated for the three progressive steps on the base case profile when filtering synthetic feed with the WFMF membrane. The obtained gradients were compared to the gradients calculated when using a flux enhancement strategy. A successful flux enhancement strategy should have a lower gradient. Also the gradients were used to assess the effectiveness of cleaning/removing of the particles that restricts flux. The gradient of a clean WFMF membrane should be high at the initial stages of filtration. The equation below was used to calculate flux decline in the membrane.

$$\text{Gradient (flux decline)} = \frac{J_2 - J_1}{t_2 - t_1} \qquad \textbf{3-1}$$

Where, J_2 is the flux taken at point 2 of the declining slope of flux and point J_1 is initial point or the proceeding point in the flux decline slope measured in LMH. t_2 is time 2 corresponding to J_2 and t_1 was the initial time corresponding to J_1.

3.4.7 Overall gradient

In filtration, the path of filtration is important; however, the overall gradient of pure water flux at constant pressure is zero because a horizontal line is formed. The overall gradient for a highly fouling feed which has an almost vertical slope is infinity. It was on these two extremes that the overall gradient assessment of flux enhancement strategies was used. The best flux enhancement strategy was expected to a have a gradient that approaches the gradient of pure water in relation to the worst performing flux enhancement strategy. Overall, the gradient was also used to assess the effectiveness of cleaning or flux recovery. A clean membrane is expected to have a high gradient because of high filtration rate; this became an effective tool to assess the effectiveness of backwashing and brushing.

3.5 Flux enhancement strategies

In this section various flux enhancement strategies were studied on their effectiveness of enhancing high fluxes on the WFMF membrane. All the flux enhancement experiments were repeated three times for repeatability and replicability. The reported figures in this work are averages.

The performance of each flux enhancement strategy was measured against the base case. For each flux enhancement experiment the base case fouling profile was plotted. The base case profile indicates the lowest flux possible for a specific feed, WFMF membrane and operating conditions. Two external membranes in the membrane pack were used to plot the base case curve. These two membranes were isolated from any flux enhancement strategy. The further away the flux was from the base case curve when employing a specific flux enhancement strategy, the more it was deemed successful. The larger the difference between the base case flux and the flux obtained with any of the flux enhancement strategies meant better performance. The differences were calculated on a daily basis as well as an overall flux after the experiments.

$$\% \text{ flux improvement} = \frac{J_0 - J_1}{J_0} \times 100 \qquad \textbf{3-2}$$

Where J_0 is the average flux for the base case (no flux enhancement is employed) in LMH, J_1 is the average flux for the enhanced flux in LMH.

3.5.1 Effect of backwashing

Backwashing is the reverse of filtration. The permeate collected in the permeate tank was used to backwash the membrane. It was possible to select the membrane module that was to be backwashed because of the individual module isolation valve. During backwashing, the backwashing water level in the backwashing tank was kept constant by level controller LC1. The isolation valve of the membrane module being backwashed was shut. This was to ensure that filtration on that specific membrane had been stopped and that the filtration fluid did not flow out through this path. Once the bottom isolation valve was shut, the backwashing valve at the top of the module was opened and the backwashing was started, (see Fig. 3-4). Reverse filtration only begins once the inside (permeate side) of the WFMF membrane is full of liquid and the liquid is forced out through the membrane pores into the feed side. The backwashing duration was 15 minutes. When it was completed, the backwashing valve was shut and the permeate valve was opened to drain for a duration of 5 minutes. This was to ensure that the WFMF membrane was free of all backwashing permeate before normal filtration recommenced. Immediately after normal filtration had begun, flux measurements were carried out to evaluate the effectiveness of the backwash and turbidity test was also done to ensure that backwashing did not damage the membrane. Backwashing cycles were varied in 3, 6 and 12 hour cycles (frequency).

3.5.2 Effect of brushing

WFMF membrane brushing was done in-situ and the duration of brushing was 1 minute per membrane side. WFMF membrane brushing was done by inserting a bottle brush in between the membranes. Contact was made with only the membrane module being targeted for brushing. Membrane brushing involved applying pressure by the brush on the membrane being brushed and the brush was moved randomly in all directions. During membrane brushing, filtration on that specific membrane was stopped by shutting the isolation valve at the bottom of the membrane module. Upon completion of one side of the membrane module, the next was brushed as well. Brushing cycles were varied at frequencies of 3, 6 and 12 hours and the brushing duration was maintained at 60 seconds per module side (120 seconds per module).

3.5.3 Effect of aeration

Aeration in submerged membrane systems is intended to create turbulence at the membrane surface which controls/minimizes the accumulation of fouling particles on the membrane surface. The membrane scouring air was supplied by the air blower as shown in Figure 3-4. The air flow rate was varied as 10, 20 and 30 ℓ/min to study the effect of scouring rate on fouling minimization/flux enhancement. The aeration rate reported was per module. The perforations on the air diffuser were 0.5 mm and the membrane spacing was 0.5 cm. Permeate flow controller (V2) was used to manipulate/change the air flow rate to the air diffuser, (see Figure 3-3). Aeration was continuous throughout the filtration period at a steady air flow rate. Baffles in between the membranes were inserted to ensure that air does not escape to the space where the bubbles will not make contact with the membrane. The starting point of the aeration rate was taken from literature available data which is based on the economically used values in MBRs (Eurombra, 2006; Kazemzadeh-Afshar *et al*, 2012)

3.5.4 Effect of reduced permeate flow

High instantaneous flux (initial flux at the beginning of filtration) is common in constant pressure operated membrane systems. A high instantaneous flux drives the filtration process to be above critical flux, resulting in a high fouling rate and a decline in flux. This experiment was conducted to study the effect of reducing/restricting permeate flux such that the filtration process was operated at a sub-critical flux (where little/no deposition on the membrane surface occurs). Permeate flux was controlled manually (by hand) by controlling valve V2. V2 was adjusted such that the permeate flux from the membrane pack was stable at 10 LMH. After some time of operation when the flux dropped below 10 LMH, V2 was opened to ensure that a stable flux of 10 LMH was obtained from the membrane pack. Figure 3-4 shows a schematic diagram for the process.

3.5.5 Pre-coating the membrane

The interaction between the foulants and membrane is a determining factor of the extent of membrane fouling. To study the effects of membrane interaction with foulants, (prior to filtration of the synthetic feed), big diameter $Ca(OH)_2$ solution was first filtered through WFMF membrane. 2 g/ℓ $Ca(OH)_2$ solution was prepared and filtered through the membrane pack. The particle size of the $Ca(OH)_2$ that was used is 2 µm. The filtration was allowed to run for 1 hour allowing the calcium particles to accumulate on the membrane surface. Filtration of $Ca(OH)_2$ was conducted in the filtration system as shown in Figure 3-3. After an hour, the filtration tank was drained-off and $Ca(OH)_2$ solution and synthetic feeds were introduced to the filtration tank. Care was taken that turbulence was not created as the synthetic feed was introduced such that the calcium layer on the membrane surface was not removed. The experiment was conducted to study the effect of pre-coating WFMF membranes with coarse $Ca(OH)_2$ prior to filtration of synthetic feed.

3.6 Optimization of the flux enhancement

Selected flux enhancement strategies were optimized using different optimization methods. In the aeration experiment, the aeration rate was increased from 10 L/min to, 20 and 30 L/min. This was to evaluate the effect of aeration rate on flux decline. Backwashing optimization was done by increasing and reducing backwashing duration time. Backwashing time was varied between 10, 20 and 30 minutes. Brushing was optimized in a similar way with backwashing where the brushing time was reduced and increased in order to evaluate its impact on flux. Brushing time was reduced from 60 seconds to 30 seconds.

Also, selected flux enhancement strategies were combined together in order to study the effect of combined flux enhancement strategies. Backwashing was combined with brushing as well as with aeration. The results of the optimization were recorded and compared to the un-optimized strategies.

3.7 Cleaning and assessment of cleaning strategies

The reason for membrane cleaning is to recover the flux of the fouled membrane to its original flux. Membrane cleaning is an integral part of any membrane system. It usually involves stopping of all filtration and doing a thorough cleaning of the membranes. The cleaning could be hydraulic, chemical or both. Cleaning methods are typically supplied by the membrane suppliers, with the aim of recovering flux to that of a new membrane or at worst to 90% of its original flux. To clean the WFMF membranes, different commercially available cleaning strategies were used. During WFMF membrane cleaning, filtration was stopped. Each individual membrane was removed from the pack and was cleaned solely. Cleaning strategies used for the WFMF membranes were:-

- Rigorous brushing of the membranes in tap water.
- Rigorous brushing of the membranes in a commercial soap solution followed by rinsing with tap water.
- Soaking the membranes in a 0.1% NaOCl solution for 1 hour followed by mild brushing of the membrane and rinsing in tap water
- High pressure water cleaning using a Karcher high pressure cleaner. The model of the high pressure cleaner was; HD 6/16-4 M Plus at 30 bar, 230 l/h and 25°C.
- Soaking of the membranes in a 0.1% NaOCl solution and rinsing with tap water.

The test criteria for the effectiveness of cleaning were the physical observation of the mechanical and integrity of the membrane after cleaning. Flux recovery was evaluated in relation to the original flux of a new membrane and separation performance maintained after cleaning. After membranes were cleaned, a PWF experiment of the cleaned WFMF membranes was done and the results were compared to their original PWFs of a new membrane. The difference between the PWF of a new WFMF membrane and cleaned WFMF indicated the success of the membrane cleaning method. A successful cleaning method is that with little or no difference between the twoPWFs.

3.7.1 Mechanical integrity strength and separation performance after cleaning test

The mechanical and integrity strength of the membrane after cleaning was done by visually inspecting the membranes after every cleaning and identifying leak point that would have developed as a result of cleaning. This was done for each cleaning strategy. A damaged membrane was expected to have higher fluxes than normal and the quality measured by turbidity was expected to deteriorate. To test for damage membranes, PWF and turbidity tests were done after every cleaning exercise.

Chapter-4 RESULTS AND DISCUSSIONS

This chapter consist of results and discussion on the development and evaluation of flux enhancement and cleaning strategies for the woven fibre microfiltration membrane. This section is divided into 6 parts, WFMF membrane pore size determination, base layer determination, evaluation of flux enhancement strategies, comparison of the strategies, and evaluation and optimization of the cleaning strategies. The results are compared to a base case for flux enhancement and pure water fluxes for cleaning.

Statistics

All the results presented in this section are averages of at least two repeated experiments, this was done to test and show the reproducibility of the results and give confidence on them. The study that was conducted in this work was an experimental study, therefore accuracy and repeatability of each experiment was important, this was tested by means of standard deviation. The standard deviation was done on the entire flux enhancement and cleaning strategies experiments and the average standard deviation on all the experiments was less than 1.

4.1 WFMF membrane characteristics determination

The purpose of the experiment was to determine the pore size of WFMF membrane by inferring its pore size to a known filter media pore size that has similar turbidity rejection capacity. Table 4-1 shows the results obtained when 0.8 µm size $Ca(OH)_2$, solution of 0.5 g/ℓ was dissolved in de-ionized and was filtered with different pore sizes filter paper and then with WFMF membrane.

Table 4-1 Turbidity rejection of a 0.8 µm solution by different mediums

Filter Media	**Lime feed turbidity (NTU)**	**De-ionized water turbidity (NTU)**	**Filtrate turbidity after 0 sec (NTU)**	**Filtrate turbidity after 1 min (NTU)**	**Filtrate turbidity after 2 min (NTU)**
1 µm Filter paper	10	0.12	7.3	4.1	1.01
0.45 µm Filter paper	10	0.12	0.51	0.32	0.28
0.2 µm Filter paper	10	0.11	0.12	0.11	0.13
WFMF Membrane	10	0.12	0.48	0.39	0.24

The results in Table 4-1 show that the WFMF membrane's turbidity rejection capacity is similar to a 0.45 µm filter paper. Even though the average particle size of the $Ca(OH)_2$ used was 0.8 µm, there could be larger particles and even smaller particles. It could be seen from Table 4-1 that the WFMF membrane had a similar rejection capacity with a 0.45 µm filter paper. Filtrate turbidity after 0 seconds was checked as the first filtrate sample. This sample represents the true rejection capacity of the filter or the membrane, because there was little or no additional secondary layer that had developed on the filter media at the initial stage of filtration.

As filtration time increased from 60 to 120 seconds, the rejection capacity for 1 µm filter paper, 0.45 µm filter paper and WFMF membrane improved. This indicated that a secondary layer on the filter media had developed that helped to improve the rejection capacities of these filters. On the 1 µm filter paper almost all the particles that are responsible for turbidity passed through the filter paper causing filtrate turbidity to be 7.3 NTU. This result meant that the majority of the particles in the solution were less than 1 µm in diameter, therefore they passed through the filter paper. But more than 90% of these particles were rejected by WFMF membrane but not all were rejected as it was the case with 0.2 µm filter paper. The conclusion from this analysis was that the rejection capacity of the WFMF membrane was better than 1 µm filter paper but less than the 0.2 µm filter paper.

The close proximity in rejection capacity between the WFMF membrane and 0.45 µm filter paper lead to the conclusion that WFMF membrane pore size is 0.45 µm or very close.

4.2 Membrane fouling

Section 4.2 focuses on the fouling of the WFMF membrane, examined was the feed characteristics, flux, and membrane material.

4.2.1 Fouling characteristic of different feeds

The purpose of the experiment was to demonstrate the effects of different feed samples on the fouling of the WFMF membrane. Feed samples with different foulants were filtered through the WFMF membrane and the results are shown in Figure 4-1.

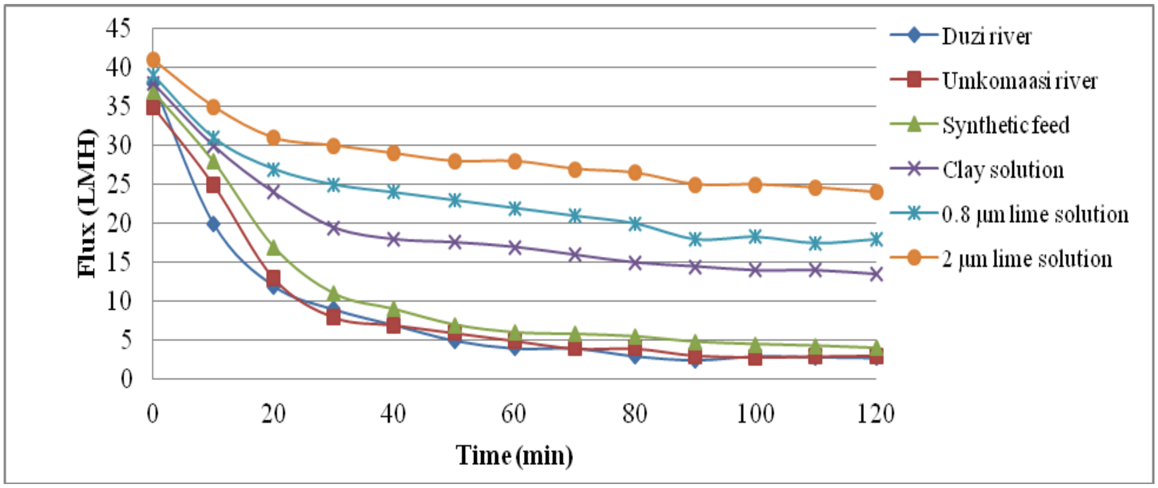

Figure 4-1 Fouling profile for different feeds

4.2.2 Fouling profile

Figure 4-1 shows the flux profiles obtained during filtration of six different samples, Duzi River, Umkomaasi River, synthetic feed, 3.5 g/L of clay solution, 3.5 g/L of 0.8 µm lime solution and 3.5 g/L of 2 µm lime solution. The results show that three samples had the highest flux decline, these were Duzi River, Umkomaasi River and synthetic feed. The 2 µm lime solution had the lowest decline in flux followed by 0.8 µm lime solution and lastly clay solution. The three samples with the highest flux decline rate had microbial components which was absent in the other feed samples. The high rate of fouling was therefore due to these microbial components. The microbiological components as shown in Table 3-1 were represented as total coliforms and *E.coli*. The samples with a lower decline in flux were single component solutions of lime and clay. The differences amongst these single component samples were the particle sizes of the components, which were 0.8 µm lime, 2 µm lime and ± 0.45 µm clay. The clay particle size is discussed in the next section.

It is also observed from Figure 4-1 that the flux decline in all six samples progressed in three distinctive steps, particularly for the three highly fouling samples. There was a sharp decline for the first 20 minutes which was followed by a short medium decline which also lasted for 20 minutes and the last step was a slow decline which continued for the rest of the filtration period. Yoon (2011) describes these steps as the deposition and compaction steps, cake layer development and pseudo steady state. The rate of flux decline during these three steps was assessed by the gradient line and the results are shown in Table 4-2.

Table 4-2 Gradient assessment of flux decline during WFMF membrane filtration

Progressive fouling steps	**Duzi river**	**Umkomaasi river**	**Synthetic feed**	**Clay solution**	**0.8 µm lime solution**	**2 µm lime solution**
Deposition gradient (LHM/min)	-1.30	-1.10	-1.01	-0.70	-0.60	-0.50
Cake layer development gradient (LMH/min)	-0.25	-0.30	-0.40	-0.30	-0.15	-0.10
Pseudo steady state gradient (LMH/min)	-0.05	-0.05	-0.06	-0.06	-0.08	-0.06
Overall gradient (LMH/min)	-0.29	-0.29	-0.28	-0.20	-0.17	-0.12

Table 4-2 shows obtained gradients for the different feed samples at different filtration stages. The results shown in Table 4-2 confirm the trend shown in Figure 4-1. The highest decline in flux for all the samples occurred in the first 20 minutes (deposition and compaction step). The multi-component feed samples (Duzi, Umkomaasi and Synthetic feeds) had a flux decline rate which was greater than 1 LMH/min. This lasted only for 20 minutes, while the single component feed samples had a flux decline rate during the same period which was less than 1 LMH/min. The interesting observation from the results in Table 4-2 is the close proximity of the cake layer development step gradient and the overall gradient. The overall gradient tells the fouling rate of the sample on a WFMF membrane for the entire duration of the experiment. The similarities of the overall gradient and the cake development gradient could mean that the big gradient during deposition and compaction is offset by the small gradient during the pseudo steady state stage.

To assess whether there was any cake development during the deposition and compaction stage of filtration, which could contribute to the high flux decline during this stage, the WFMF membrane during synthetic feed filtration was taken out of the filtration tank to observe the membrane surface. Figure 4-2 shows a visual representation of the WFMF membrane in the first 20 minutes of filtration. Figure 4-3 shows a visual representation of the WFMF membrane after the entire filtration period.

Figure 4-2 Picture of WFMF membrane after 20 minutes of filtration

Figure 4-3 Picture of WFMF membrane for the entire period

Figure 4-2 shows that very little development of the cake layer occurred on the WFMF membrane during the first 20 minutes of filtration when it is compared to the fully developed cake layer on the WFMF membrane as shown in Figure 4-3. The observation in Figure 4-2 implies that deposition on the WFMF membrane that resulted in a high decline in flux could have occurred on the WFMF membrane internals rather than on the surface.

4.2.3 Feed sample characterisation

It was not possible to characterise Duzi and Umkomaasi Rivers beyond the observed characteristics of the two rivers shown in Table 3-1. However, the synthetic feed was prepared to have the same fouling characteristics as these two rivers and this was achieved (see fouling profile in Figure 4-1). The characteristics of the synthetic feed were inferred from the two rivers. According to Bessiere *et al.* (2009), Nguyen *et al.* (2012) and Abdelrasoul *et al.* (2013), microbiological, bacterial and organic foulants mostly foul membranes by attaching on the membrane surface and adsorbing onto the membrane surface. The results shown in Table 4-3 reveals the effect of adsorption fouling on the WFMF membrane. This experiment was conducted by soaking the WFMF membrane in the synthetic feed for 5 minutes without any filtration taking place. After the membranes were soaked in the synthetic feed, a pure water flux (PWF) was done and compared to the pure water flux before the immersion experiment.

Table 4-3 Adsorption test of synthetic feed foulants on a WFMF membrane

Test Variable	**Flux 1 (LMH)**	**Flux 2 (LMH)**	**Flux 3 (LMH)**	**Average flux**
Pure Water Flux before soaking WFMF membrane in synthetic feed	61	58	59	59.3
Pure water flux after 20 minutes soaking of WFMF membrane in synthetic feed	43	45	39	44

Results in Table 4-3 shows that soaking the WFMF membrane in a synthetic feed definitely resulted in a loss in PWF. The loss in PWF after soaking the WFMF membrane was attributed to adsorption fouling because no filtration was allowed to occur during this experiment to avoid deposition on the membrane. The conclusion from these results was that the synthetic feed sample had components that adsorbed on the WFMF membrane. These components were believed to be from the activated sludge that was added into the clay solution and these are represented as Total coliforms and *E.coli* on the feed characteristic table in Table 3-1. The same behaviour after soaking was expected for the samples from Duzi and Umkomaasi Rivers, because of the similarities in these feed samples. The soaking experiment was not conducted for the single component samples because in these samples there were no organic, bacterial or microbiological foulants present in these feed samples.

Particle size in relation to the membrane pore size affects the fouling rate on the membrane. Colloidal matter has a higher fouling rate than suspended particles (Lee *et al.*, 2004; Hwang *et al.*, 2008). The lime particle size was known because it had already been determined by the supplier as explained in **Section 3.2.1** and was used to determine the particle size of clay. Table 4-4 shows the results obtained when the clay particle size was determined using the MSE method.

Table 4-4 Clay particle diameter determination results using the MSE method

Filter Media	**Clay solution turbidity (NTU)**	**Filtrate turbidity after 0 sec (NTU)**	**Filtrate turbidity after 1 min (NTU)**	**Filtrate turbidity after 2 min (NTU)**
1 µm Filter paper	10	8.23	6.4	3.1
0.45 µm Filter paper	10	5.1	2.54	1.95
0.2 µm Filter paper	10	0.2	0.18	0.18
WFMF Membrane	10	4.85	3.51	1.4

Filtrate turbidity at zero filtration time represents the true filter separation capacity. This turbidity is before a cake layer developed on the surface of the filter paper which would help to improve the separation capacity of the filter paper. On the 1 µm filter paper, almost all the particles that cause turbidity passed through the filter paper. This was concluded that the clay particles in a solution are less than 1 µm in diameter. When same solution was passed through a 0.45 µm filter paper, almost 50% of the turbidity causing particles passed through the filter paper. This inferred that about 50% of the clay particles are less than 0.45 µm in diameter. The 0.2 µm rejected all the turbidity. It was interpreted as all the clay particles were bigger than 0.2 µm in diameter. From the results shown in Table 4-4 it was concluded that the clay particle size distribution is ± 0.45 µm in diameter.

4.2.4 Membrane material

The WFMF membrane was made of a hydrophobic polyester fabric and the membrane pore size was concluded to be 0.45 µm and the thickness of the membrane material was 1.2 mm. The visual magnified microscopic view of a WFMF membrane is shown in Figure 4-7.

A difference in the fouling rate/flux decline rate was observed amongst the single component feed samples. The major difference amongst these feed samples was mainly particle sizes. The 2 µm lime feed sample had the biggest particle size amongst the single component feed samples and the fouling rate for this feed sample was the least. When the particle size decreased to 0.8 µm and to 0.4 µm the fouling rate increased. The reason for the increased fouling rate with a decrease in particle size was attributed to pore plugging and narrowing. This was deduced from the fact that the WFMF membrane had a pore size of 0.45 µm and the clay particles were found to be averaging 0.4 µm. This means that these particles went into the pores and got stuck in the pore channel thereby increasing the hydraulic demands from the system.

The high hydraulic demands resulted in flux reduction because the filtration was operated under constant pressure. The particles getting stuck in the pore channel of the WFMF membrane was also facilitated by the fact that WFMF membrane had an unconventional thickness of 1.2 mm. This is much higher than the average industrial membrane thickness which is less than 0.5 mm. In most or all filtration processes, larger particles in the feed samples cannot get into the membrane pores to block it. They are only deposited on the surface of the membrane. Furthermore, because the size of the packing density is low, the cake layer formed will be porous. The hydraulic demand by a porous cake layer are a lot less than the hydraulic demands of multiple plugged membrane pores due to the compactness of the cake layer formed. This explains the differences in fouling rates of feed samples that had varying particle sizes dispersed in the feed sample.

To further confirm if the dispersed particles in the feed solution did not get trapped on the membrane surface but rather went through the pore channels and fouled the membrane internally, the WFMF membrane was immersed in the feed solution and was taken out after 20 minutes of filtration and a very small layer was seen on the membrane surface as shown in Figure 4-2.

Figure 4.4 shows a profile of turbidity removal by the WFMF membrane, however visual observation of the membrane is shown in Figure 4-2, does not show any particles on the membrane surface. This demonstrated that clay particles entered into the WFMF membrane pore and fouled the membrane internally by pore blocking and constriction. Striemer *et al.* (2006) found that thick membranes are highly fouled by colloidal particles.

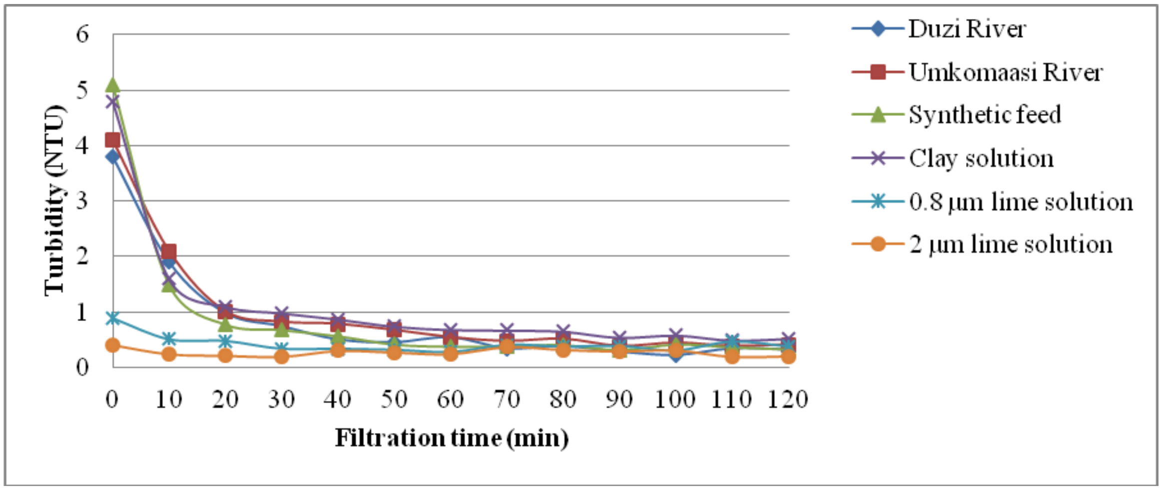

Figure 4-4 Turbidity profile for different filtrate from different feed samples

The synthetic feed sample fouled the membrane in two ways, by adsorption as seen from the results in Table 4-3 and pore plugging by small diameter particles that enter the pore area of the membrane. This explains the fouling differences between multi-component feed solutions and single component feed samples.

The high gradient during the first 20 minutes of filtration as seen in Figure 4-1 and Table 4-2 was attributed to a high instantaneous flux. A high instantaneous flux occurs on a clean membrane because at this stage of filtration the hydraulic filtration demands are minimal and are only to drive the liquid through the membrane pore area. The head pressure drives the feed and its particles towards the membrane at a high rate, this is because of the minimum hydraulic demand on a clean membrane (not fouled membrane). The high filtration rate moves the filtration process to be above critical flux which then leads to high fouling rates and flux decline. Therefore, the high fouling rate during the first 20 minutes of filtration is due to operating the membrane process above its critical flux. This type of operation is notorious for high fouling rate which is in agreement with Lee *et al.*, 2004 and Hwang *et al.*, 2008 who found that during the initial stages of filtration high instantaneous flux cause severe fouling on the membrane. Because the filtration rate is too high as a result fouling particles a dragged more rapidly towards the membrane surface due to high velocity through the membrane pores.

With the multi-component feed samples adsorption fouling was another factor that made these samples have a high fouling rate than single component samples. Adsorption fouling does not need filtration to be in progress to start. All that is required for adsorption fouling to start is contact between the adsorptive components and the membrane material. The WFMF membrane is hydrophobic and adsorption fouling is severe on hydrophobic membrane materials. It was also was found by Flemming and Schaule (1988) that hydrophobic membrane material have a high affinity to bacterial or biological attachment or adsorption.

4.3 Flux enhancement strategies

The flux enhancement and cleaning strategy experiments were done only with the synthetic feed. For each experiment, a base-line fouling curve was done in conjunction with the flux enhancement strategy used for comparison between the flux enhancement strategy used and the fouling curve.

4.3.1 Effect of aeration on flux

An inherent nature of all filtration systems is the accumulation of rejected particles by the filtering media due to size exclusion. The accumulated particles increase the hydraulic requirements for filtration thereby resulting in a reduction in flux. In this section the effects of air scouring on the WFMF membrane was investigated in order to maintain a high flux. The results are shown in Figure 4-5. The aeration rate reported was per module.

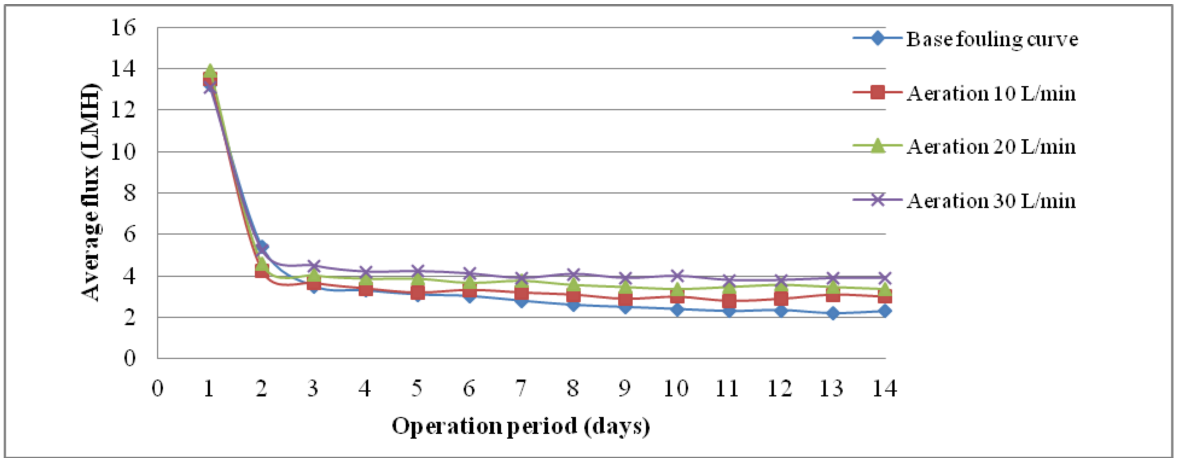

Figure 4-5 Effect of aeration on flux enhancement

Figure 4-5 shows the obtained trends when the WFMF membrane was aerated at different air flow rates during synthetic feed filtration. The graph shows that air scouring of the WFMF membrane improves its flux. Increasing the aeration rate had a positive impact on flux. Aeration rate of 10 L/min.m^2 improved the flux by 11%; aeration of 20 and 30 L/min had an improvement of 25% and 36% respectively. The average flux improvement was calculated using Equation 3-2, where the improvement is from the base case average to the enhanced flux average.

The gradient/rate of flux decline for the period of 14 days was 0.29 LMH/day without aeration. When the WFMF membrane was aerated at a rate of 10 L/min, the gradient was reduced to 0.23 LMH/day; this was further improved to 0.21 and 0.20 LHM/day for 20 and 30 L/min respectively.

Figure 4-6 shows the fouled membranes as seen visually on the effect of aeration rate. A thick slimy cake layer is seen on the surface of the membrane without aeration and the thickness and the visibility of the cake layer is reduced as aeration rate is increased.

Figure 4-6 Photos of fouled membranes at different aeration rates

The reason for improved flux when aeration rate is increased lies in the effectiveness to remove the accumulated particles from the membrane surface. The aeration rate of 30 L/min is more effective for removing the accumulated particles on the membrane surface than the aeration rate of 10 L/min. This is because for the high aeration rate of 30 L/min that there was no significantly visible cake layer on the surface of the membrane when compared to the membrane aerated at a rate of 10 L/min. Therefore, the high aeration rate of 30 L/min is more effective in controlling and even eliminating cake build-up on the WFMF membrane, but the success only contributes to a maximum of 36% in flux improvement. A flux improvement of only 36% at a high aeration of 30 L/min was not expected because much higher fluxes have been reported in MBR systems. However, this could be explained by the differences in particle sizes in the feed solutions. Particle sizes of the flocs in MBRs can be up to 20 times larger than the membrane pore size.

The magnified photo of the WFMF membrane is shown in Figure 4-. This amongst other reasons can help to explain the reason for only 36% improvement in flux at such a high aeration rate.

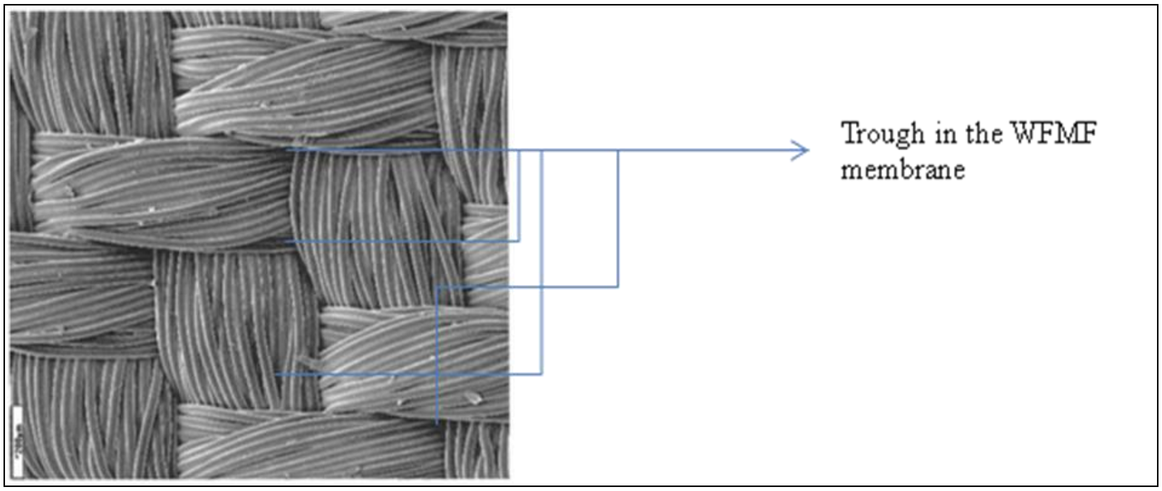

Figure 4-7 Magnified microscopic view of the WFMF membrane (X1000)

Figure 4-7 shows that WFMF membrane is not a flat-smooth membrane, but has trenches and troughs as the fibres are twisted and weaved into a uniform pattern. It is in these trenches that the expected fine particles in a solution get stuck and block the membrane pores and results in a decline in flux, even at high aeration rates. Even in-between the fibres, the fine particles are expected to get in and hinder permeate flow. The air bubbles from air scouring cannot get to these trenches and whether the fine air bubbles produced during aeration do not get into the trenches or not, they do not have the necessary force to remove the trapped clay particles.

In Section 4.2.1 it was discovered that the clay particles were averaging 0.4 µm in diameter against the membrane pore size of about 0.45 µm and a thickness of 1.2 mm. The size of the clay particles in relation to the WFMF membrane pore size is similar. It is therefore credible that the particles could get into the membrane pores and get stuck inside the pore channel and trenches. Air bubbles cannot impact these particles. They therefore block permeate path and the net result is a reduction in flux. A cross-flow velocity is not effective in dispersing fine particles away from the membrane surface and eventually into the membrane pore. This is a common finding amongst membrane researchers such as Song and Menachem Eiimelech (1995); Hong *et al.* (1997); Le-Clech *et al.* (2003);

Striemer *et al.* (2006); Field *et al. (*2010); Hai and Yamamoto (2011) and Pradhan *et al.*(2012).

It was also discovered in Section 4.2.1 that the WFMF membrane is prone to adsorption fouling with the synthetic feed. This occurs on contact between the macromolecular protein-like foulants present in the feed solution. Aeration was not effective in controlling this type of fouling.

Air scoring is effective in controlling the build-up of a cake layer on the surface of the WFMF membrane at a maximum achievable aeration rate of 30 L/min. Controlling the cake build-up contributes to 36% improvement flux. This finding implied that there are other fouling mechanisms that cause a decline in flux on the WFMF membrane other than cake layer build-up on the membrane surface which cannot be controlled by air scouring. The results in Table 4-3 and Table 4-4 lead to a conclusion that the WFMF membrane fouls by pore blocking and constriction and adsorption fouling. A combination of these two fouling mechanisms contributed to high fouling rate/high flux decline observed on the WFMF membrane.

4.3.2 Effect of backwashing frequency on flux

There is a tendency of particles that are smaller than the pore size of the membrane to penetrate the pore and get stuck inside the membrane pore. Backwashing is esentially a reversal of filtration and is intended to remove the stuck particles in the membrane pore thereby improving the flux through the membrane. An experiment to study the effect of backwashing of the WFMF membrane was conducted and the results are shown in Figure 4-8. Varied in the experiment was the backwashing cycle and it was varied at the intervals of 3, 6 and 12 hours.

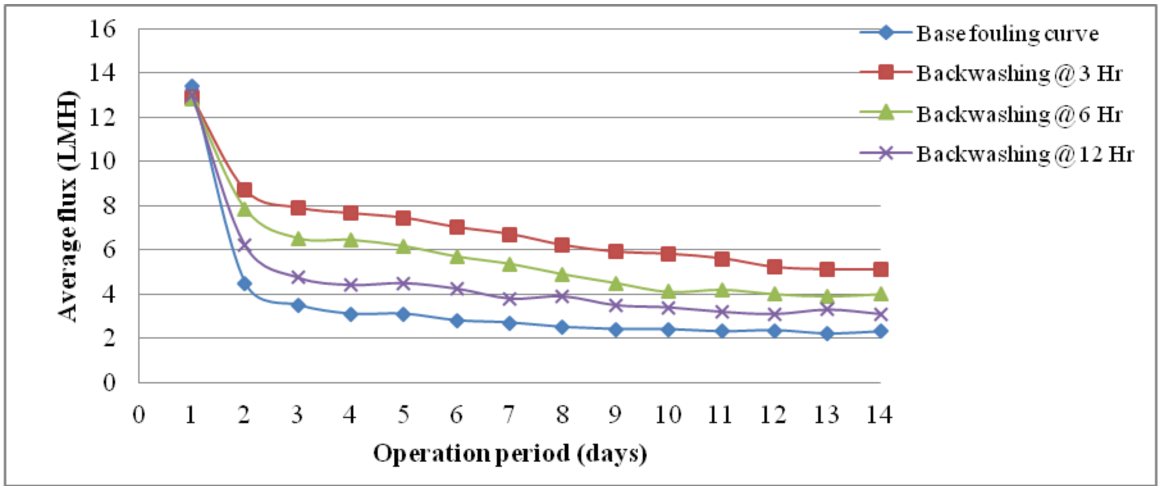

Figure 4-8 Effect of backwashing on flux enhancement

Figure 4-8 shows that backwashing improves the average flux when compared to the absence of backwashing. It can be seen that when the backwashing cycle was increased from 12 to 6 and to 3 hourly and the average flux from the WFMF membrane was improved. The average flux was improved by 30.2%, 69.5% and 121% for 12, 6 and 3 hour cycles of backwashing respectively. Backwashing removes or clears the membrane pore channel of any particles that could have been stuck on the membrane thereby obstructing or even blocking the path of the permeate. The high flux improvement by backwashing implies that particles were getting trapped in the pore channel and therefore narrowing the membrane pore which led to flux decline.

Every backwashing sequence is intended to recover flux to almost the membrane's original flux when cleaned. Figure 4-9 shows the extent to which various backwashing cycles recovered flux.

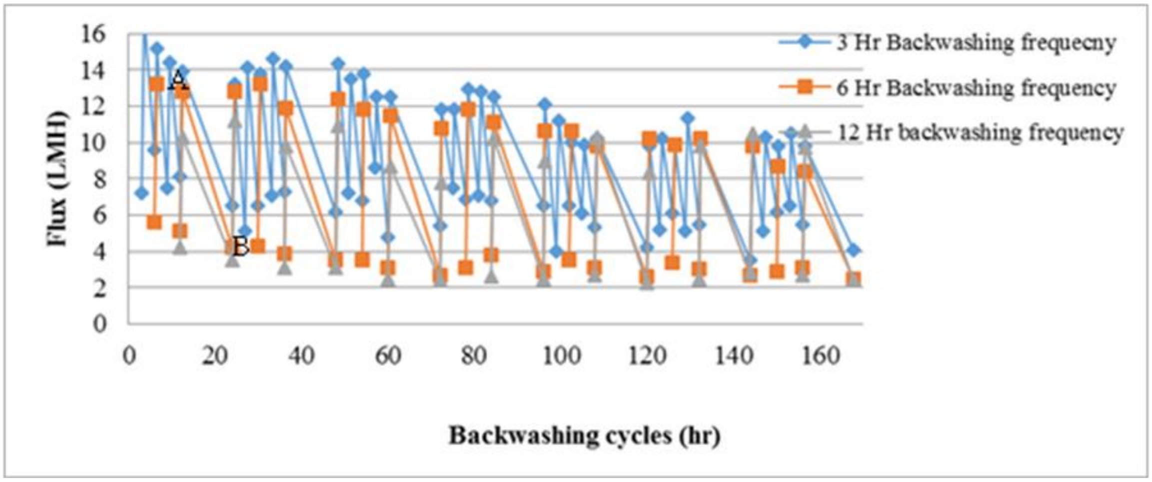

Figure 4-9 Profile for backwashing and flux recovery

In Figure 4-9 it can be seen that high backwashing cycles had consistently high flux recovery in the first few hours. As the filtration time for backwashing continued for the three cycles, the trend observed was becoming similar. The highest flux recovery was in the 3 hour cycle followed by the 6 hour cycle.

Thick membranes like WFMF membranes are prone to internal fouling and backwashing is mostly the method used to mitigate against the full establishment of internal fouling on a membrane. Figure 4-8 shows that the particles trapped in the membrane pores and trenches of WFMF membrane earlier shown in Figure 4-7 were removed successfully by backwashing. The factor that made backwashing successful for flux improvement in the WFMF membrane was due to the fact that there was a direct contact of the backwashing water with the particles that blocked the permeate flow area. Also, the backwashing water was supplied at a pressure of 1 kPa higher than the filtration pressure. This helped to remove the trapped particles in the membrane pore area.

The high frequency of backwashing resulted in high recovered flux. This was attributed to the fact that with higher frequency the particles trapped in the pore areas and trenches were not allowed to strongly attach in the pore, because these were frequently being loosened and removed by the higher pressure backwashing water. Whereas, with longer cycles the particles are stuck in the membrane pores and are allowed to acclimatise and afterwards are then trapped in the pore thereby needing a greater pressure and more time to remove.

A limitation of backwashing is that the particles removed/loosened in the membrane pore are quickly brought back into the pore area as soon as filtration commences again. A high slope gradient is an indicator of this phenomenon. The gradient was calculated for the three backwashing cycles and the results are shown in Table 4-5 below.

Table 4-5 Effect of backwashing on flux recovery, base flux, lost flux and gradient

Backwashing frequency (hr)	Average Recovery Flux (LMH)	Average Base Flux (LMH)	Average gradient per backwashing cycle
3	12.2	6.2	-2.03
6	10.1	3.45	-1.45
12	9.7	2.8	-1.41

A high frequency backwashing cycle of 3 hours had the largest gradient of -2.03 LMH/min while the other two cycles had similar gradients of 1.41 and 1.45 LHM/min. The average gradient per backwashing cycle was calculated within the single cycle as shown by A and B in Figure 4-9 and the equation used was Equation 3-1. This result was expected because the 3 hour cycle had a high flux recovery. This means that when the filtration recommenced, the high recovered flux resulted in high instantaneous flux. High instantaneous flux results in a high fouling rate because of the high fluid velocity through the membrane pores, which increases the particle drag force towards the membrane surface and eventually into the pores. However, during backwashing the particles are not dragged from the bulk solution but are loosely positioned on the membrane surface by the backwashing effect, so these particles are quickly brought back into the pore area when filtration begins and quickly plugs the membrane pores. This is the reason why high frequency backwashing had a high gradient.

In Figure 4-9 it can be seen that towards the later part of filtration the gradient and flux recovery for the 3 hour cycle dropped significantly. This was attributed to irreversible fouling settling in on the membrane and was believed to be driven by high operating fluxes that were observed with the 3 hour backwashing frequency.

The impact of backwashing is only in the membrane pore area because once the backwashing water leaves the pore area to the feed side it loses its pressure and becomes equal to bulk feed pressure due to the large feed area. Therefore, backwashing is only effective in the WFMF membrane internals (pore area) and not so much on the surface of the membrane. The 121% improvement in flux when the 3 hour backwashing cycle was used was therefore attributed to the removal of particles hindering permeate flow to the filtrate side and hence, resulting in flux decline.

The declining slope of the backwashing average flux profile in Figure 4-8 and Figure 4-9 indicate that irreversible fouling might be occurring on the membrane and backwashing which periodically produces high instantaneous flux increased the rate of the formation of irreversible fouling. Backwashing by nature has no impact on surface fouling of the membrane and is not rigorous enough to deal with adsorption fouling on the WFMF membrane.

4.3.3 Effect of in-situ brushing

In-situ brushing is not common in submerged systems but can be a practical flux enhancement strategy for small scale systems. This section looks at the effect of brushing the WFMF membranes in-situ in between filtration cycles on flux improvement. Each WFMF membrane was brushed for 1 minute by inserting the brush in between the membrane. The brushing cycle was varied between 3, 6 and 12 hours and the results are shown in Figure 4-10.

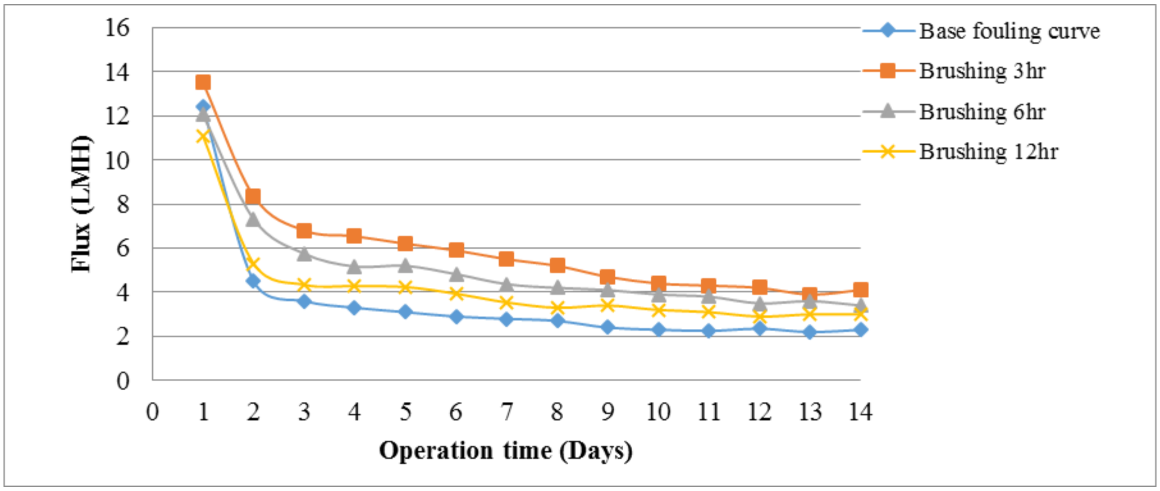

Figure 4-10 Effect of In-situ brushing on flux enhancement

Figure 4-10 shows the results of varying the brushing cycle on flux improvement for the WFMF membrane. Clearly visible is that when the brushing frequency is increased, the average flux also increases. The improvement in flux was calculated to be 87.3%, 55.9% and 28.7% for 3, 6 and 12 hour cycle of WFMF membrane brushing respectively. The WFMF membrane brushing was intended to remove the fouling layer on the membrane surface. The improvement in flux implies that periodic brushing of the WFMF membrane had an impact on flux although it was for a short time.

Figure 4-11 shows flux recovery after every brushing sequence. It can be seen that there was not much difference in the recovered flux between the 3 and 6 hour brushing cycles. The 12 hour cycles brushing had a little lower flux recovery than the other two at start but towards the end of the filtration period the flux recovery for all three cycles were similar. The reason for the improvement in the flux at high frequency of brushing was attributed to the high recovery flux and quick brushing before the flux dropped too low for the 3 hour brushing cycle. This helped maintain a high average filtration flux for both 3 and 6 hour brushing cycles.

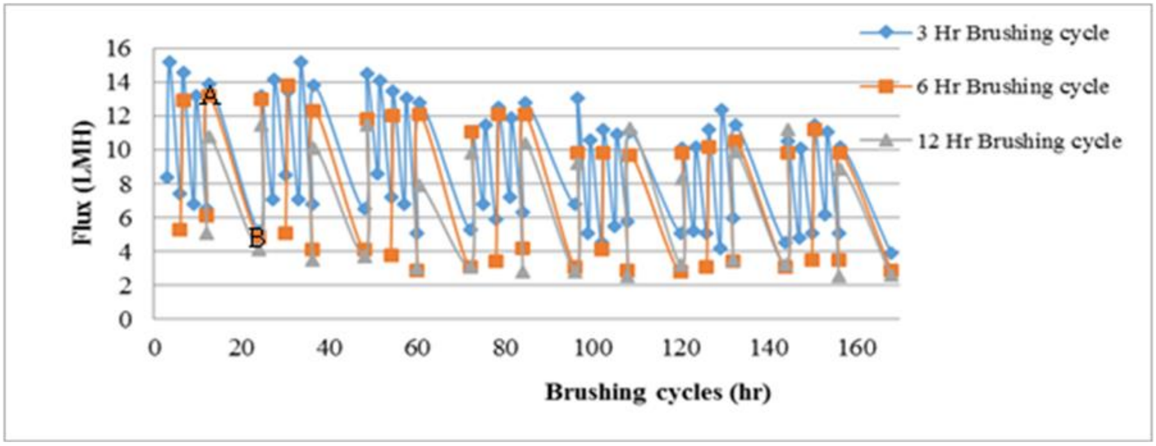

Figure 4-11 In-situ brushing profile and flux recovery

To assess how long the recovered flux by brushing was maintained, a gradient on each of the brushing cycles was done. High gradients would mean the high fluxes are short lived before fouling settles in again. The results are tabled in Table 4-6.

Table 4-6 Effect of backwashing on flux recovery, base flux, lost flux and gradient

Brushing frequency (hr)	Average Recovery Flux (LMH)	Average Base Flux (LMH)	Average gradient per brushing cycle
3	11.72	6.1	-2.78
6	11.35	3.8	-2.87
12	10.1	3.2	-3.2

The high gradients seen in Table 4-6 attest to the fact that the fouling layer on the WFMF membrane was being removed, but it was quickly re-establishing again on the membrane surface. The average gradient per backwashing cycle was calculated within the single cycle as shown by A and B in Figure 4-11 and the equation used was Equation 3-1. In Section 4.2.1 and Figure 4-1 it was shown that particle deposition on the WFMF membrane took about 20 minutes on a clean membrane and the gradient was found to be around 1.4 LMH/min. Particle deposition started once filtration had recommenced and was the reason for the high gradient as seen in Table 4-6. This sudden decline in flux was then attributed to adsorption fouling. This was derived from the fact that the fouling on the WFMF membrane after brushing was re-established too quickly according to the results in Table 4-6.

Adsorption fouling occurs on contact with adsorptive materials, so when the cake layer on the membrane surface was removed by brushing, the adsorptive materials gained access to the WFMF membrane surface which was originally shielded by the cake layer. This explains the high gradients after membrane brushing. This is in agreement with a study by Field *et al.* (2010).

Even though membrane brushing was not continuous, a direct contact was made between the membrane and the brush with much force applied. This then made the shear achieved during brushing to be greater. As a result a high instantaneous flux after brushing resulted in high average fluxes. Membrane brushing had access to the trenches on the surface of the WFMF membrane. This also helped remove hidden fouling layers in the trenches which would have contributed to the membrane filtration area loss during filtration if not removed.

Periodic brushing helps in de-stabilising the adsorption fouling on the WFMF membrane. This instability created on the adsorption fouling helps to loosen the fouling making the membrane permeable to fluid flow and the net result was the improvement in flux. The decline in the average flux profile obtained during periodic brushing is an indicator that irreversible fouling could be growing on the WFMF membrane and could be as a result of periodically exposing the membrane to adsorption fouling after every brushing.

4.3.4 Controlled flux operation below critical flux

High filtration flux is believed to lead to high membrane fouling. In this section, the permeate flux was restricted to 10 LMH from the start of filtration. The purpose of the experiment was to stop the high instantaneous flux that occurs at the beginning of filtration because the WFMF membrane was clean and hopefully maintained the flux at 10 LMH for prolonged periods. Figure 4-12 shows the obtained results when permeate flux was restricted to 10 LMH.

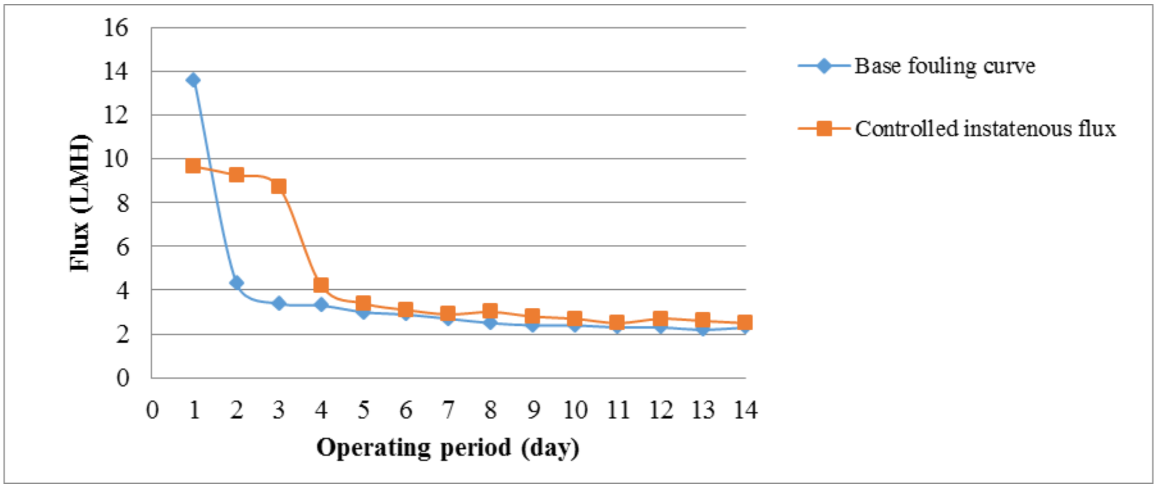

Figure 4-12 Effect of controlled flux on fouling

It can be seen from Figure 4-12 that restricting/controlling filtration at 10 LMH delayed the sharp decline in permeate flux as seen on the base fouling curve. However, after 3 days a similar decline in flux as with the base fouling curve was observed. Controlling permeate flux at 10 LMH was intended to eliminate the effects of high instantaneous flux at the beginning of filtration by reducing the drag force that transports particles from the bulk solution to the membrane surface.

The typical starting flux at time zero for the base fouling curve was 35 LMH against 10 LMH for the controlled flux profile. The sudden decline in flux after 3 days implies that there was a sudden rise in fouling of the WFMF membrane after this period but this was not observed in the base fouling curve. This was unexpected and could not be due to sudden changes in the feed or operating conditions because the sudden drop was not seen in the base fouling curve which was subjected to the same conditions. The reason for the sudden decline in flux of the WFMF membrane was believed to be due to the way the flux from the WFMF membrane pack was controlled.

The flux was controlled by manipulating the discharge/permeate flow V2, (see Figure 3-4). The limitation with this type of control was that the WFMF membrane module is hollow on the permeate side. Therefore, the permeate valve V2 could be completely shut yet filtration would be proceeding on the membrane surface. Also the WFMF membrane bulges and expands when the permeate side of the membrane is filled with water, expanding its capacity to accumulate liquid on the permeate side. This means that the restrictions that were done on the permeate via V2 had no effect on the filtration process taking place on the membrane surface. The implication of this is that normal filtration as with base fouling curve was occurring on the membrane surface and the membrane was getting fouled. The permeate was accumulating on the membrane permeate side because of the restrictions of V2, hence during the first few days of filtration the constant flux of 10 LMH was the accumulated permeate on the membrane permeate side. The sudden decline in flux after 3 days was because the accumulated volume in the permeate side had run out because the incoming permeate from the membrane had dropped due to fouling. So the decline in flux shown on the third day shows that the membrane was fouled and this fouling was believed to have occurred on day one but was hidden by the constant flux that was being drawn out of the membrane.

Controlling the filtration flux for WFMF membrane should be done on the feed side by reducing the driving pressure, because if flux control is done on the permeate side, the design of the WFMF membrane allows filtration to proceed even when the permeate valve is shut.

The profile of the base fouling curve and controlled flux after 5 days had similar fluxes. This implied that flux control in the filtrate side did not mitigate fouling of the WFMF membrane. The sharp decline after 3 days is a further indicator of pre-existing fouling even before that day. The results of this section were not compared to the rest of the flux enhancement strategy results, because proper flux control was not achieved. The decision not to further investigate the flux control on the feed side was because of the monitoring requirements that would have been required. For example, locating and moving the level float to a new position and moving it back up for the rest of the experiment. This exercise would have required more than one filtration tank, which was not the case. Also, 24 hour monitoring of flux would have been necessary in-order to determine exactly when the time that flux started dropping to below 10 LMH for proper analysis and conclusion from the experiment. The latter also was not possible.

4.3.5 Comparison of flux enhancement strategies

The flux enhancement strategies were compared against each other. The comparison was to identify the flux improvement strategy that resulted in the highest flux improvement and possibly optimize them. Figure 4-13 shows the bar chart comparison of the strategies.

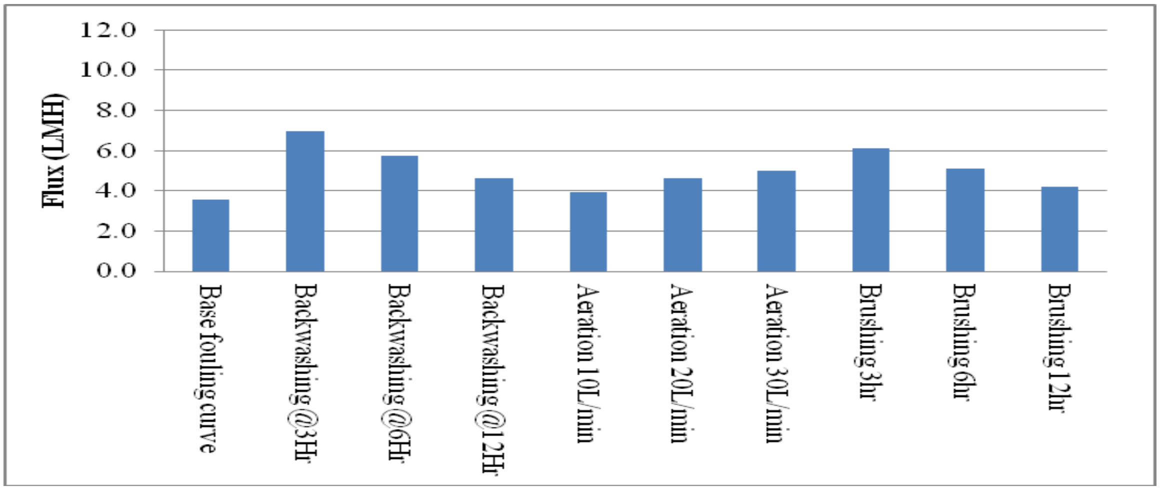

Figure 4-13 Comparison of flux enhancement strategies

From Figure 4-13, it can be seen that the backwashing cycle of 3 hours resulted in the highest average flux. This result implies that significant fouling of the WFMF membrane was from particles plugging the pores of the WFMF membrane and regular backwashing of the membrane was successful in removing these particles. The removal of the particles from the pore area cleared the path for the permeate to flow freely, resulting in high flux.

Second to the 3 hour cycle backwashing was the 3 hour cycle brushing, brushing forcibly removes the particles that were stuck on the WFMF membrane trenches. It was believed adsorption fouling was also loosened due to the force applied in brushing, even though this was for a short period of time. Removal of the particles in the membrane trenches, and agitating the adsorption fouling layer on the membrane was believed to be the main contributors to the good success of the 3 hourly brushing.

Flux improvement by controlling permeates flux and air scouring at 30 L/min was less than expected for both the flux enhancement strategies. The WFMF module design and the way flux was controlled made the flux control strategy ineffective because filtration rate on the membrane surface where fouling actually takes place was not impacted by the way the flux enhancement strategy was implemented. The lack of success by aeration was attributed to the fact that the shear offered by the rising air bubbles was not strong enough to remove particles trapped on difficult areas on the WFMF membrane.

4.3.6 Fouling test

Fouling resistance can be hydraulically or chemically removed. The sign of fouling on a membrane is a loss of PWF when compared to the original pure water flux. At the end of the filtration cycle on each of the flux enhancement strategies, a PWF test was carried out just before the membranes were cleaned. The purpose of the experiment was to determine the presence of fouling. The results are shown in Figure 4-14.

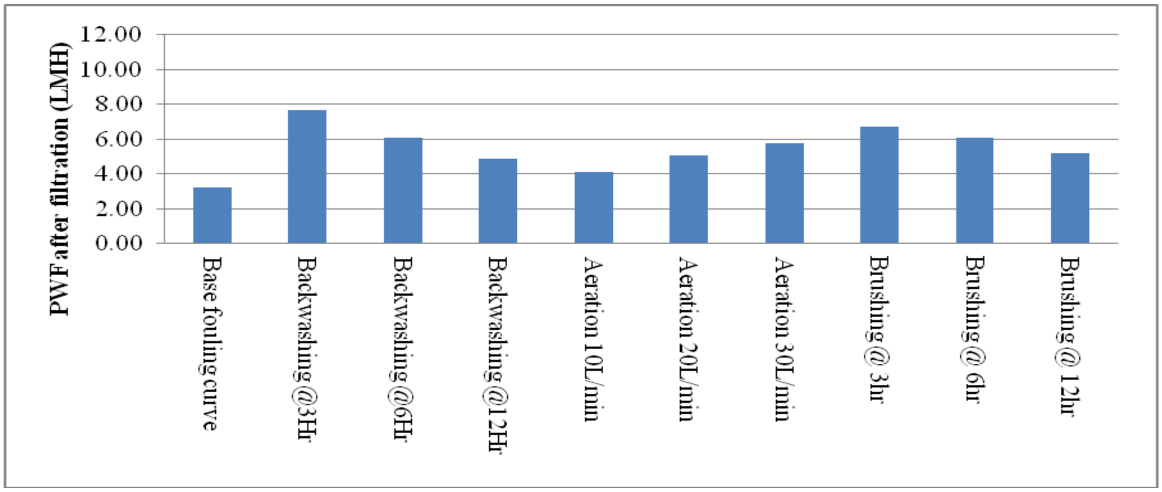

Figure 4-14 Fouling test results for various flux enhancement strategies (PWF)

The results shown in Figure 4-14 are very close to those in Figure 4-13. The results shows that flux enhancement strategies that were successful in maintaining high fluxes also resulted in less irreversible fouling. The results suggested that periodic and continuous agitation of the WFMF membrane minimized the full establishment of irreversible fouling on the WFMF membrane. PWF results conducted on the dirty membranes had a slightly higher flux than at the final flux at which the membrane was stopped. This was expected because when the membranes were rinsed before the PWF run, the loose cake layer which also adds some resistance to filtration was removed. Also during PWF run there was no further addition of fouling on the membrane surface.

4.4 Optimization of flux enhancement strategies

Flux enhancement optimization was done on the following strategies: 3 hour cycle backwashing, 3 hour cycle brushing and membrane pre-coat. The first two strategies were selected based on the good performance in flux enhancement as shown in Figure 4-13.

4.4.1 Backwashing optimization

Backwashing effectiveness is affected by its frequency and duration. The effect of changing the backwashing duration on a 3 hour cycle of backwashing was studied in this section and the results are shown in Figure 4-15.

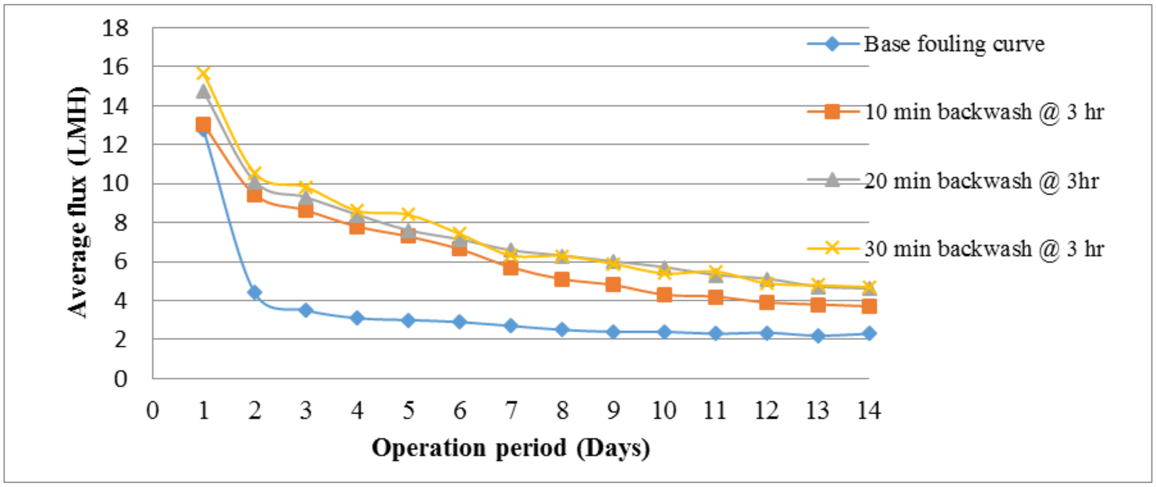

Figure 4-15 Backwash optimization profile

In Figure 4-15, it can be seen that when the duration of backwashing is increased from 10 minutes to 20 minutes there is an improvement in the flux decline profile. The improvement in the flux from the 10 minutes to 20 minutes backwashing was 15%. However, when the backwashing duration was increased to 30 minutes, no significant improvement is seen. The flux improvement was only 6.8%. The results suggest that a 20 minute backwashing time for a 3 hour backwashing cycle was the best backwashing duration for the WFMF membrane. This is true because the average flux for 15 minutes backwashing time was 6.94 LMH against 7.25 LHM for 20 minutes backwashing time.

Two main reasons could be used to explain the decline in average flux and low flux profile when the backwashing duration was reduced to 10 minutes. The first reason could be that the WFMF membrane permeate side needs to be completely filled with water before the actual backwashing starts. The second reason could be that there was insufficient time to remove the strongly attached particles on the membrane pore area when the backwashing time was reduced to 10 minutes. In Section 4.3.2, the average flux for 15 minutes backwashing duration at a frequency of 3 hour cycle was 6.94 LMH, but in Section 4.3.3 where the duration was increased to 20 minutes for the same backwashing frequency and the average flux was increased to 7.25 LMH. Therefore, it can be concluded that backwashing in Section 4.3.2 could be optimised by increasing the backwashing duration.

Figure 4-16 shows the effect of backwashing duration on irreversible fouling resistance. Irreversible fouling resistance as explained in Section 4.2.6 was determined by running a PWF after the filtration period where different flux enhancement strategies were used.

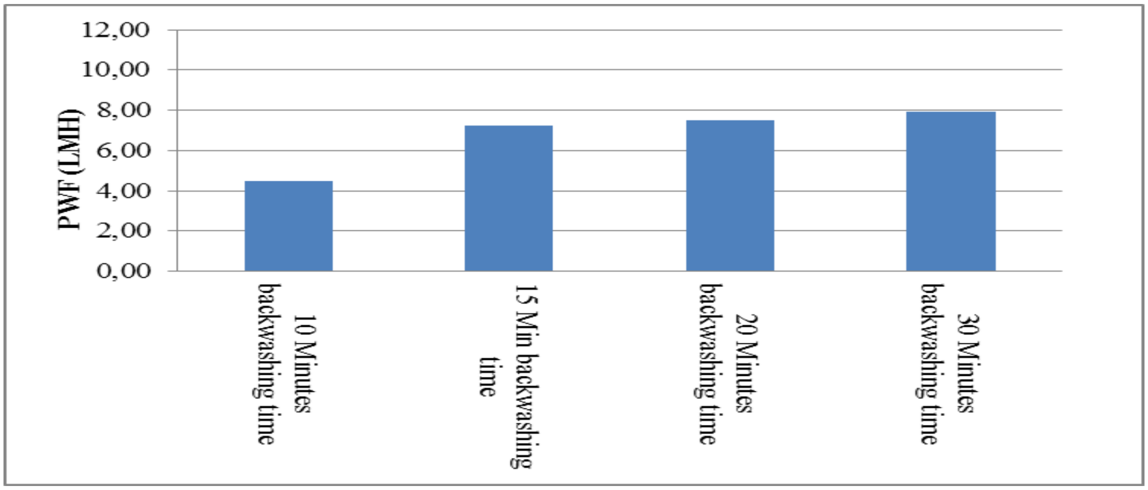

Figure 4-16 Backwashing duration effect on irreversible fouling at 3 hour interval

Figure 4-16 compares the effectiveness of the backwashing duration to avoid irreversible fouling. From the above figure it can be seen clearly that when the duration of backwashing was increased pure water flux increased. Higher pure water flux after filtration was interpreted as low irreversible fouling on the WFMF membrane. The pure water fluxes obtained were 4.5, 7.23, 7.5 and 7.9 LMH for 10 15, 20 and 30 minutes backwashing duration time respectively.

4.4.2 In-situ brushing optimization

To optimise membrane in-situ brushing, the duration of WFMF membrane brushing was studied. The brushing time was varied but the frequency was kept constant. The duration of in-situ brushing was varied at 20, 30 and 60 seconds. The purpose of the study was to investigate the effect of brushing duration on flux decline control. The obtained results are plotted in Figure 4-17.

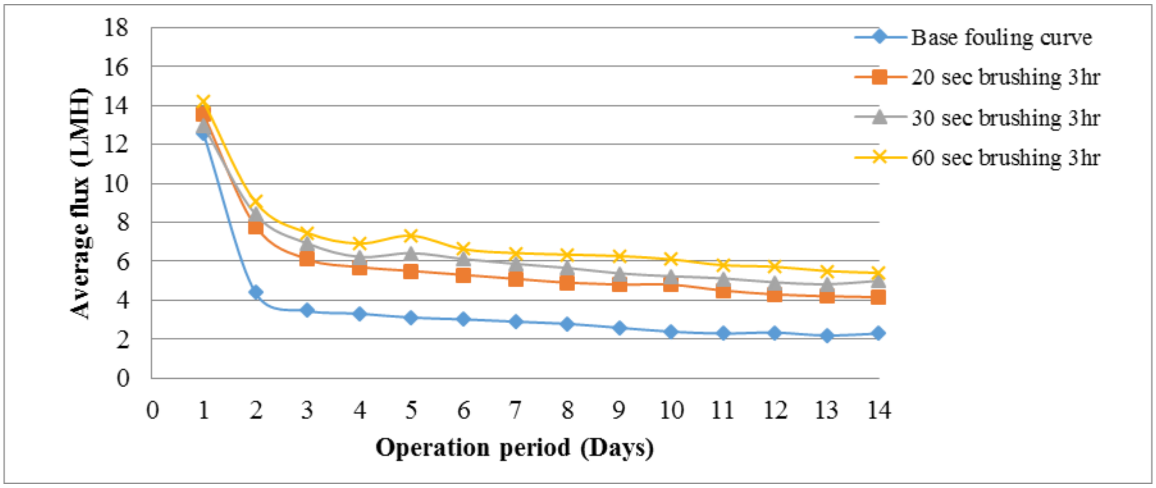

Figure 4-17 In-situ brushing optimization profile at 3 hour intervals

Figure 4-17 shows that when the brushing time is increased from 20 seconds to 30 second and finally to 60 seconds per module side, the flux decline is reduced and higher flux averages are obtained. The increase in average flux reduction in the flux decline with the brushing time was observed to be due to the following reasons. Firstly, the longer brushing time means that sufficient time was given to remove attached particles on the membrane surface and also more filtration area was brushed. Brushing loosens the agglomerated particles due to the van de Waal forces. These reasons were believed to be the cause of the increase in average flux when the brushing time is increased. Therefore, 60 second brushing used in Section 4.2.3 was found to be the best brushing time for the WFMF membrane. Increasing the brushing above 60 seconds was not studied because of its impracticality for larger systems with numerous membranes.

The effect of increasing the brushing duration to mitigate against fouling was studied, and the results are shown in Figure 4-18.

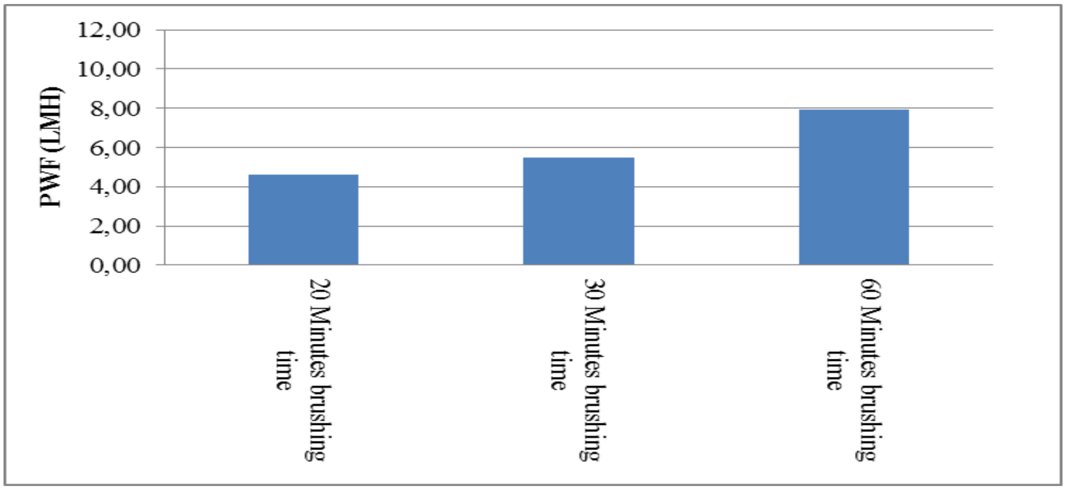

Figure 4-18 In-situ brushing duration effect on fouling at 3 hour intervals

The results shown in Figure 4-18 shows the effect of increasing the brushing time from 20 seconds to 30 and 60 seconds per module side on fouling mitigation. The results show that in all three occasions by increasing the brushing time, fouling is reduced further.

4.4.3 WFMF membrane pre-treatment

In Section 4.21, it was seen that filtration of 2 µm $Ca(OH)_2$ solution did not result in large decline in the flux that would be related to adsorption fouling or WFMF membrane internal fouling. It was therefore decided to filter first a 2 µm $Ca(OH)_2$ solution first for 30 minutes in order to coat the WFMF membrane surface with the 2 µm $Ca(OH)_2$ particles. The results of the experiment are plotted in Figure 4-19.

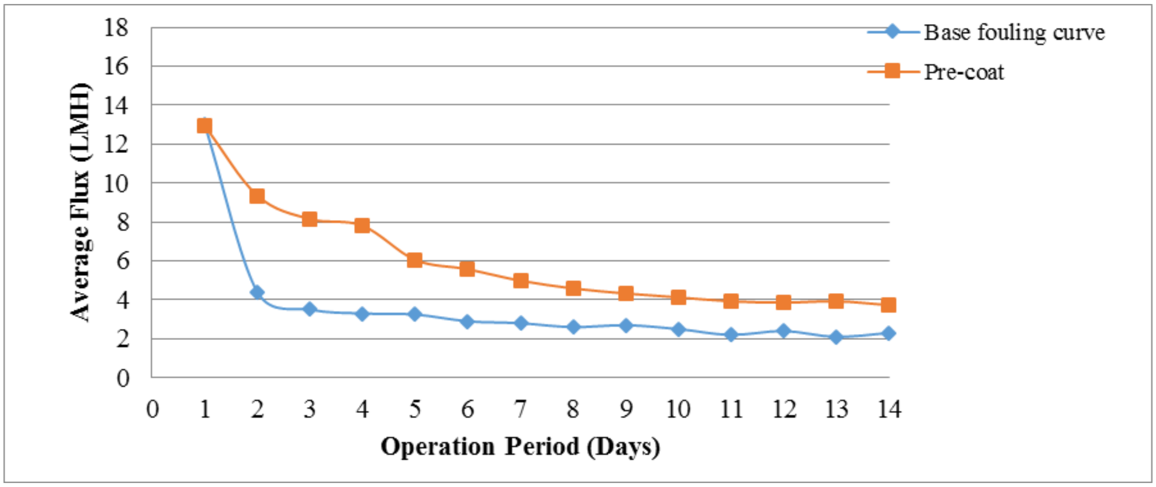

Figure 4-19 Effect of pre-coating the membrane with 2 µm Ca(OH)2 solution

Figure 4-19 shows the results of the effect of pre-coating the WFMF membrane with 2 µm $Ca(OH)_2$ particles prior to filtration of synthetic feed. The results show a major improvement from the base fouling curve when pre-coating of the WFMF membrane was done. The average improvement in flux from the base fouling case was 66.2%. The large decline in the first day to the second day was reduced by a factor of 2.4. The flux decline gradient for the base fouling curve from day one to day two was calculated to be 8.6 LMH/day and the pre-coated membranes gradient dropped to 3.6 LMH/day. The reduction of flux decline and the improvement in the average value flux was attributed to the following reasons.

Firstly, the 2 µm $Ca(OH)_2$ particles were bigger than the WFMF membrane pores therefore these particles were porously packed on the membrane surface. The colloidal particles from the clay solution during filtration had no access to the membrane surface because of the $Ca(OH)_2$ particles that were already coated on the membrane surface. This prevented WFMF pore narrowing and plugging by the colloidal clay particles. WFMF membrane pore plugging and narrowing by the colloidal clay particles in the synthetic feed was believed to be the major cause for sharp decline in the filtration flux in the first day of filtration. This finding was in agreement with many other researchers such as Hong *et al.* (1997), Striemer *et al.* (2006) and Hwang *et al.* (2008).

In Section 4.2.1, it was also discovered that the WFMF membrane was prone to adsorption fouling by the synthetic feed. Adsorption fouling occurs upon contact between the adsoptive material in the feed solution with the membrane surface. Coating the WFMF membrane with 2 µm $Ca(OH)_2$ particles prevented contact between the membrane and the adsorptive foulants in the feed solution. Coating the WFMF membrane with 2 µm $Ca(OH)_2$ particles was effective in mitigating adsorption fouling, hence the decline in flux in the first day of filtration was minimal when compared to the base case.

4.4.4 Backwashing combined with aeration plus brushing

The WFMF membrane was found to be prone to both internal and external fouling, therefore, flux enhancement strategies that only deals with either internal fouling or external fouling could not be completely effective. In this section, the effect of combining backwashing which mostly deals with internal fouling, was combined with the external fouling control strategies, brushing and aeration. The results of the study are plotted in Figure 4-20.

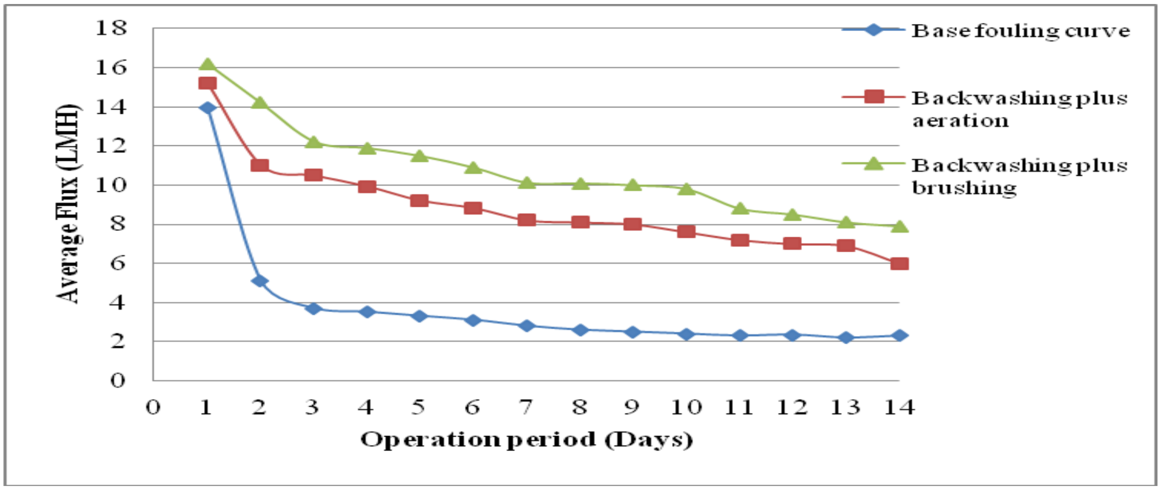

Figure 4-20 Effect of integrating 3 hourly 20 minutes backwashing with aeration and 3 hourly 60 second in-situ brushing

Figure 4-20 shows the results of combining backwashing with either aeration or brushing. The results shown indicates that combining 3 hours cycle backwashing with 3 hour cycle brushing yielded the best flux enhancement. Backwashing was conducted for a duration of 20 minutes and the brushing was conducted for a duration of 60 seconds. The overall flux decline gradient for this combination (backwashing and brushing) was the slightly higher than that of backwashing and aeration, this was merely because of high initial flux in the former combination. The overall gradient for backwashing plus aeration was $0.70LM^2H$/day and the overall gradient for backwashing plus brushing was $0.71LM^2H$/day. The small gradient indicated a good flux enhancement strategy by these combinations. The improvement in flux the in relation to the base-case, when backwashing was combined with brushing, was 187.2%. This was the highest of all the flux enhancement strategies.

The flux profile of combining backwashing with 20 $L/min.m^2$ continuous aeration is also shown in Figure 4-20. Good success with this combination is also evident. The average improvement in relation to the base-case in flux when backwashing was combined with aeration was 135.7%. This implies that the fouling control is best tackled by a combination of strategies rather than single flux enhancement strategies. In a combination approach, the weakness of the one strategy is covered by the strength of the other. Backwashing is weak in tackling membrane surface fouling but is very effective in controlling internal fouling.

Backwashing was found to be ineffective in surface fouling in Section 4.2.2. This means that particles from the membrane internals (pore channel) were brought to the surface of the membrane/pore entry. When filtration recommenced, the particles brought to the membrane surface/pore entry quickly are brought back into the membrane pore. These particles were not swept away from the membrane pore and this resulted in high flux decline gradients calculated from Figure 4-8 results. When backwashing was combined with either aeration or brushing, these particles brought to the membrane surface were dispersed back into the bulk solution away from the membrane surface. The combination of flux enhancement strategies slowed down the quick re-entry of particles into the membrane pore. This resulted in the high flux improvement and small flux decline gradients calculated from Figure 4-20 results. The reason backwashing combined with brushing gave better results than when combined with aeration was due to the fact that brushing offered stronger shear and could access more difficult areas that air bubbles from aeration could not get to.

4.5 Cleaning strategy performance

Fouling is inherent in all membrane systems, and is manifested by the loss of the PWF from the original PWF of clean/new membrane. Membrane cleaning aims to recover the membrane's original PWF after cleaning. In this section several membrane cleaning strategies were used to clean the WFMF membrane to recover its original flux after synthetic feed filtration. The cleaning strategies that were used to clean the WFMF membrane were brushing in tap water, brushing in a soap solution, brushing in a 0.3% NaOCl solution, soaking in a 0.3% NaOCl solution and high pressure cleaning. The results of PWF after cleaning are shown in Table 4-8.

Table 4-7 Flux recovery table of comparison

	New mem.	**External brushing with feed**	**Brush.+ soap**	**Soak in 0.3% NaOCl + brushing**	**Soak in 0.3% NaOCl only**	**High press. clean**	**Fouled mem.**
Pure water flux (LMH)	60	43	49	56	47	58	3
Per. recovery		71.7	81.7	93.3	78	97	5

Table 4-7 shows the results of flux recovery by different cleaning methods of a WFMF membrane. The membranes were cleaned using various cleaning methods as shown in Table 4-7. Once the membranes were cleaned the PWF experiments were repeated.

External brushing of the WFMF membranes with tap water recovered the PWF to 71.7%. The 71.7% recovery of flux by brushing was based on the removal of external fouling on the WFMF membrane because brushing had no access to the internal fouling of the WFMF membrane. External brushing of WFMF membranes removes both deposited particles on the membrane surface, trenches and also the adsorbed fouling on the membrane surface.

Brushing the membrane with a commercial soap solution recovered the PWF by 81.7%. The use of a detergent with brushing ensured that the strongly attached foulants on the membrane surface were loosened and removed by brushing. The emulsifying and dispersion effect of detergents were believed to be the factor that caused brushing of WFMF membrane in a commercial soap solution more effective than brushing the membrane with tap water alone. This is in agreement with a study by Abdelrasoul *et al.* (2013) which found that detergents are good emulsifying, dispersion and surface conditioners particularly for mud and clay stains.

Soaking of the WFMF membrane for an hour in a 0.3% hypochlorite solution followed with brushing recovered the PWF to 93.3%. Soaking the membranes only in the same solution recovered only the 78% of the PWF. The result shows that hypochlorite is effective in removing/loosening the strongly attached foulants both on the membrane surface and membrane internals. The loosened/removed particles included strong hydraulically attached particles on the membrane surface and chemically attached foulants (adsorbed fouling). The adsorbed materials in this study include bacteria, microbiologic contaminants, and organic particles. The combination of soaking the membranes in a 0.3% NaOCl and brushing ensured that fouling on the membrane was loosened by the oxidation and disinfection mechanism of NaOCl. Brushing ensured that the loosened attachments on the membrane are removed. The combined cleaning effect with this method resulted in the high PWF recovery of 93.3%. Chemical cleaning by oxidation and disinfection have also been studied by several researchers such as Holman and Ohlinger (2007), Arnal *et al.* (2012) and Abdelrasoul *et al.* (2013) who found similar findings when NaOCl was used as the cleaning chemical.

High pressure cleaning of the WFMF membranes recovered 97% of the PWF. This type of cleaning was only mechanical. 97% pure flux recovery by mechanical cleaning meant that the fouling on the membrane was more hydraulic than chemical. The low recovery by brushing alone was attributed to insufficient mechanical cleaning force by brushing alone. High pressure cleaning water was able to penetrate the WFMF membrane pores and mechanically clean the membrane surface. This method cleaned the membrane both externally and internally. According to Nguyen *et al.* (2012) microorganisms attach strongly on surfaces and aynd this explains why it had to take high pressure cleaning to recover almost all of the PWF.

4.5.1 Comparison of flux enhancement strategies

Selected flux enhancement strategies used in this study were compared to each other. The selected strategies were the most successful strategies to prevent a sharp decline in flux due to fouling and giving high average flux for the filtration period. The results are shown in a bar chart in Figure 4-21.

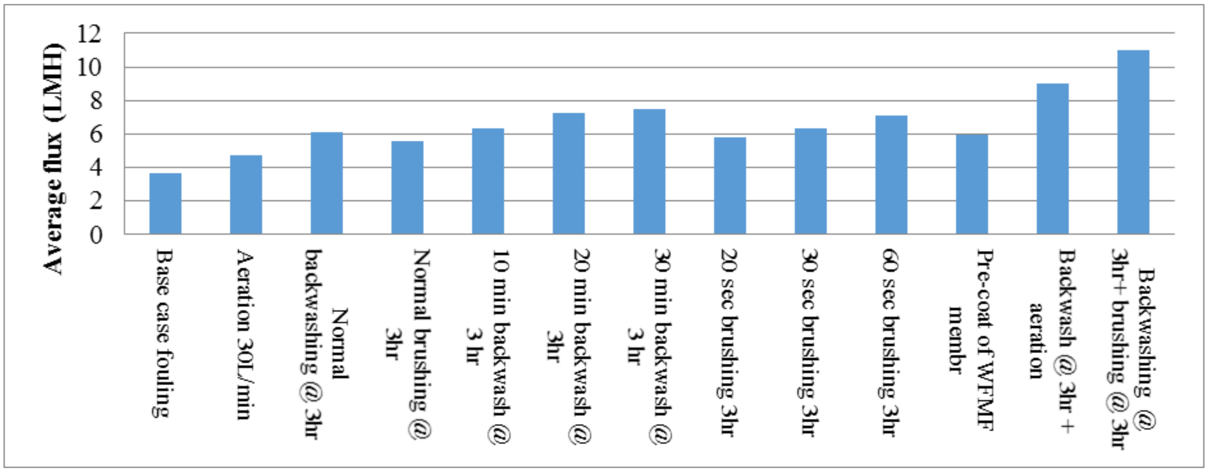

Figure 4-21 Comparison of various flux enhancement strategies on WFMF membrane

Figure 4-21 clearly displays that combining two flux enhancement strategies had the most success in mitigating against WFMF membrane fouling and yielding higher average fluxes. As explained in Section 4.3.4, the advantage of combining two flux enhancement strategies lies in the fact that the limitation of the one strategy is covered by the strength of the other and the net result is high ability of the combined flux enhancement strategy to mitigate WFMF membrane fouling.

Chapter-5 CONCLUSIONS AND RECOMMENDATIONS

The WFMF membrane is susceptible to pore plugging by colloidal material and adsorption/attachment by microbiological contaminants which take effect in the first hour of filtration. The net result is loss of more than 50% of the filtration flux in the first hour. Components such as colloidal matter that are less than 0.45 µm in diameter are responsible for pore plugging of the WFMF membrane. Microorganisms, natural organic matter and bacteria tend to foul the WFMF membrane by adsorption

High instantaneous flux after cleaning, backwashing or in-situ brushing promote WFMF membrane pore plugging by clay particles because these particles at high fluxes are dragged by the high velocity towards the membrane surface. This results in smaller particles to penetrate and enter the membrane pores which get trapped in the membrane pore and plug the pore leading to flux decline.

The physical structure of the WFMF membrane (thickness, surface view and pore size) was a major factor that caused aeration to be ineffective in flux control. Aeration was effective in dispersing the cake layer and ensuring that a cake layer does not develop on the surface of the membrane. However, aeration could not prevent entry of particles into the pores and into the membrane trenches which resulted in the real loss of flux. Once the clay particles (colloidal matter) entered into those hidden areas, aeration could not get to it and remove it. Aeration is not effective in preventing fine particles from approaching the membrane surface and depositing on it and aeration cannot prevent adsorption fouling.

Backwashing of the WFMF membrane was the most successful single flux enhancement strategy because much of the fouling of the WFMF membrane was due to pore plugging and narrowing. Backwashing is effective in removing particles that have plugged the membrane pores and lead to flux decline. Backwashing cleans the WFMF membrane internally but is weak on surface/external cleaning of the WFMF membrane.

External cleaning of the WFMF membrane was found to be satisfactory with frequent brushing of the membrane. This method ensured that difficult areas to reach like, trenches were cleaned and rigorous brushing removed the strongly attached particles on the membrane surface. When backwashing was combined with brushing the average flux improvement from base-curve was 187.2% and was 135.7% when backwashing was combined with aeration at a rate of 20L/min.

Pre-coating of the WFMF membrane with 2 µm lime was the simplest and less tedious to any other flux enhancement strategy, yet this method improved flux by 66.2%. This method improved average flux by shielding the membrane from contact with adsorptive microbiological material. Lime coating on the WFMF membrane prevented the fine clay particles from entering membrane pores.

The WFMF membrane fouls both internally by pore plugging and externally by adsorption and deposition on the membrane and a single flux enhancement strategy proved insufficient to maintain high flux successfully. Combining backwashing with either aeration or external brushing gave the best results. Backwashing is effective in removing internal fouling from the WFMF membrane, while either aeration or brushing was found to be good for external fouling.

A 0.3% hypochlorite solution was found to be effective in restoring the WFMF membrane flux. This was because NaOCl is a good oxidizing and disinfection agent for the foulants that were found in the feed. The foulants were clay particles and microbiological material. Using hypochlorite with brushing increased the effectiveness of the cleaning from 71.7% recovery and 78% recovery when using brushing alone or hypochlorite solution alone, respectively to 93.3% when combined. This was because NaOCl dissolves and loosens the fouling layer on the membrane surface and brushing ensures that the loose particles were removed from the membrane surface. This method offered a double cleaning effect.

High pressure cleaning offered a more rigorous or stronger mechanical cleaning of the membrane. The fact that high pressure cleaning was able to recover flux completely (97% flux recovery) implied that the attached foulants on the membrane surface were hydraulically attached to the membrane and did not change its structure. High pressure cleaning and chemical cleaning combined with rigorous brushing implied that the WFMF membrane is mechanically strong.

A low cost WFMF membrane was found to be able to be run continuously at good fluxes with simple flux enhancement strategies like backwashing and brushing. The robustness of the membrane fabric on rough treatment like high pressure cleaning combined with chemical cleaning and brushing, makes WFMF membrane a sustainable solution for hard and developing environments. There is a scope to rest harsher but more effective cleaning and flux enhancement strategies on the WFMF membrane because a critical limit of the membrane was not reached on this study.

RECOMMENDATIONS

It was recommended from this study that in future studies, backwashing of WFMF membrane must be done with a hypochlorite solution so that backwashing can remove adsorption fouling as well.

The filtration rate should be controlled from the feed side by reducing the driving pressure rather than shutting off the filtrate valve. The filtration rate control should be done in combination with aeration. This could effectively reduce deposition of material on the membrane surface.

It is also recommended that aeration be done with suspended carriers or beads to provide positive contact with the fouling layer developed on the WFMF membrane. It is also recommended that high instantaneous fluxes at the beginning of filtration be controlled from the feed side and not the permeate side, because of the volume that accumulates inside the membrane.

This study was more of an experimental study, because of no knowledge of the response of the WFMF membrane on flux enhancement and cleaning strategies, therefore the results of this work could be used as basis for future studies. It is therefore recommended that detailed statistics be conducted in future studies where much of the work would be focused on observational studies and correlations and models would be developed.

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APPENDIX A WFMF membrane and feed solution fouling characterization

Data for fouling of WFMF membrane

Fouling is a measure of flux decline in constant pressure system and flux is calculated by:

$$J = \frac{Q}{A}$$

Q is the volumetric flow rate in litres per hour and A is area in m^2

Driving pressure for the system was 0.4m head

Table A1: Fouling Profile for Duzi River

Period (min)	Run 1 (LMH)	Run 2 (LMH)	Run3 (LMH)
0	38	39	38
10	20	22	21
20	12	14	10
30	9	8	8
40	7	8	7
50	5	6	6
60	4	5	5
70	4	4	3.8
80	3	3	2.8
90	2.5	3	3
100	3	3.1	2.5
110	2.9	3.2	2.2
120	2.8	3	2.5

Table A2: Fouling Profile for Umkomaasi River			
Period (min)	Run 1 (LMH)	Run 2 (LMH)	Run3 (LMH)
0	40	39	37
10	28	24	22
20	12	11	13
30	9	8	8
40	8	7	6
50	6	5	4
60	6	4	3.5
70	5	3.8	3.2
80	4	3.5	3
90	3.2	2.7	2.7
100	3	3.1	2.5
110	2.8	2.9	2.2
120	2.7	2.8	2.4

Table A3: Fouling Profile for Synthetic feed River			
Period (min)	Run 1 (LMH)	Run 2 (LMH)	Run3 (LMH)
0	38	39	41
10	24	27	28
20	16	15	17
30	10	12	12
40	8	7	9
50	6.5	6	8
60	6	5	7.6
70	5.8	4.3	6
80	5.5	4.1	5.5
90	4.8	3.9	5
100	4.5	3.7	4.4
110	4.3	4	4.1
120	4	3.9	4.2

Table A4: Fouling Profile for Clay solution

Period (min)	Run 1 (LMH)	Run 2 (LMH)	Run3 (LMH)
0	38	40	41
10	30	32	30.1
20	24	25	19
30	19.5	22	17
40	18	19.2	16.2
50	17.6	18.2	16.3
60	17	17.2	15
70	16	15.6	14.9
80	15	14.3	13.7
90	14.5	13.5	13.6
100	14	13.4	14
110	14	14.1	13.2
120	13.5	13.6	13

Gradient calculation

$$m = \frac{\Delta y}{\Delta x}$$

y is the flux and x is the operation time. The first gradient is calculated in the first 20 minutes of filtration, the second gradient is calculated from 20 minutes of operation to 100 minutes while the last gradient is calculated from 100 minutes to the end of operation.

Table A5: Turbidity Profile for Duzi River

Filtration time	Run 1 NTU	Run 2 NTU	Run 3 NTU
0	3.8	2.9	3.5
10	1.9	1.2	0.98
20	0.98	0.88	0.81
30	0.74	0.68	0.74
40	0.5	0.61	0.64
50	0.45	0.54	0.58
60	0.54	0.48	0.49
70	0.34	0.41	0.52
80	0.38	0.38	0.34
90	0.28	0.39	0.32
100	0.22	0.31	0.28
110	0.34	0.32	0.3
120	0.32	0.3	0.22

Table A6: Turbidity Profile for Umkomaasi River

Filtration time	Run 1 NTU	Run 2 NTU	Run 3 NTU
0	3.9	3.6	4.1
10	1.7	0.97	1.5
20	1.01	0.87	1
30	0.82	0.74	0.87
40	0.78	0.68	0.76
50	0.67	0.47	0.71
60	0.54	0.39	0.62
70	0.47	0.47	0.31
80	0.51	0.39	0.47
90	0.38	0.37	0.56
100	0.44	0.35	0.44
110	0.38	0.4	0.53
120	0.41	0.33	0.43

Table A8: Turbidity Profile for Synthetic Feed			
Filtration time	Run 1 NTU	Run 2 NTU	Run 3 NTU
0	5.1	4.9	5.2
10	1.5	1.3	2.1
20	0.78	1.1	0.9
30	0.68	0.86	0.78
40	0.57	0.61	0.71
50	0.42	0.6	0.6
60	0.38	0.55	0.5
70	0.39	0.54	0.47
80	0.41	0.48	0.48
90	0.32	0.41	0.51
100	0.42	0.37	0.43
110	0.37	0.35	0.53
120	0.33	0.32	0.52

Table A9: Turbidity Profile for Clay solution			
Filtration time	Run 1 NTU	Run 2 NTU	Run 3 NTU
0	4.8	5.3	4.8
10	1.6	1.7	1.9
20	1.1	1.2	1.1
30	0.98	0.97	0.87
40	0.87	0.88	0.74
50	0.74	0.89	0.68
60	0.68	0.78	0.45
70	0.67	0.68	0.61
80	0.65	0.57	0.52
90	0.54	0.69	0.71
100	0.58	0.55	0.62
110	0.49	0.47	0.49
120	0.52	0.54	0.57

APPENDIX B Flux enhancement strategies

Data for aeration

Air was supplied by a blower supplying air at atmospheric pressure

Table B1: Effect of 10L/min/m2 aeration rate to fouling

Period (day)	Base fouling curve	Base fouling curve	Aeration 10L/min	Aeration 10L/min
1	8.26	9.2	9.1	9.8
2	5.4	5.1	4.25	4.5
3	3.5	3.2	3.67	3.7
4	3.3	2.9	3.4	3.5
5	3.1	3	3.2	3.1
6	3.03	2.8	3.33	3.6
7	2.8	2.7	3.2	3.3
8	2.6	2.8	3.1	2.9
9	2.5	2.6	2.9	2.8
10	2.4	2.5	3	3.14
11	2.3	2.2	2.8	2.7
12	2.35	2.4	2.9	2.9
13	2.2	2.2	3.1	3.2
14	2.3	2.3	3	2.5

Table B2: Effect of 20L/min/m2 aeration rate to fouling

Period (day)	Base fouling curve	Base fouling curve	Aeration 20L/min	Aeration 20L/min
1	9.2	9.7	9.05	9.35
2	5.4	5.16	4.6	4.7
3	3.8	3.9	4.06	4.2
4	3.5	3.6	4.16	3.98
5	3.21	3.19	4.1	4
6	3.15	3.2	3.9	3.5
7	2.89	2.98	3.544	3.9
8	2.95	3.01	3.8	4.1
9	2.49	2.6	4	3.7
10	2.7	2.97	3.7	3.8
11	2.9	3.1	3.8	3.2
12	3.1	2.8	3.8	3.9
13	2.4	2.7	3.9	4.1
14	2.6	2.6	3.7	4.2

Table B3: Effect of 30L/min/m2 aeration rate to fouling

Period (day)	Base fouling curve	Base fouling curve	Aeration 30L/min	Aeration 30L/min
1	9.12	9.5	9.1	9.07
2	4.7	5.34	5.23	6.1
3	4.1	3.18	4.5	5.5
4	3.8	3.2	4.21	4.6
5	3.5	3.05	4.23	4.4
6	3.02	3.02	4.12	4.31
7	2.97	2.9	3.9	4.1
8	3.1	3.1	4.1	3.7
9	2.1	3	3.9	3.8
10	2	2.6	4.2	4.01
11	2.5	2.3	3.8	3.9
12	2.41	2.5	3.7	3.8
13	2.34	2.4	3.9	3.9
14	3.1	2.3	3.9	3.7

APPENDIX C Cleaning and assessment of cleaning strategies

Data for backwashing

Backwashing pressure was 0.5m head and permeate was used to backwash the membranes

Table C1: Effect of backwashing every 3 hours

Period (day)	Base fouling curve	Base fouling curve	Backwashing	Backwashing
1	12.74	13.01	13.34	13.2
2	4.6	4.3	9.7	8.9
3	3.7	3.6	9.4	9.2
4	3.51	3.4	8.82	8.97
5	3.4	36	7.44	8.2
6	2.9	3.01	7.8	8.1
7	2.7	2.8	6.7	7.9
8	2.6	2.78	6.2	6.2
9	2.4	2.5	5.5	5.4
10	2.35	2.33	5.8	5.1
11	2.3	2.34	5.6	4.7
12	2.35	2.4	5	5.1
13	2.2	2.7	4.8	5
14	2.3	2.1	5.1	4.8

Table C2: Effect of backwashing every 6 hours				
Period (day)	Base fouling curve	Base fouling curve	Backwashing	Backwashing
1	13	12.87	12.89	13.21
2	4.3	4.3	6.86	6.78
3	3.7	3.7	6.53	6.47
4	3.4	3	6.45	6.55
5	3.6	2.7	6.17	6.12
6	3.02	3.4	5.7	6
7	2.8	3.2	5.37	5.4
8	2.4	3.4	4.9	4
9	2.7	2.9	4.5	4.2
10	2.31	2.7	4.1	4.1
11	2.3	3.1	4.2	4.2
12	2.2	2.4	4	3.9
13	2.4	2.9	3.9	4.1
14	2.1	2.1	4	3.8

Table C3: Effect of backwashing every 12 hours				
Period (day)	Base fouling curve	Base fouling curve	Backwashing	Backwashing
1	12.78	13.4	13.3	13.2
2	4.6	5.4	5.2	4.8
3	3.8	3.4	4.75	4.5
4	3.45	3	4.42	4.2
5	3.47	3.8	4.5	4.1
6	3.01	2.8	4.25	3.98
7	3	2.7	3.8	3.7
8	3	3.7	3.9	3.65
9	2.8	2.1	3.5	3.7
10	2.3	2.01	3.4	3.4
11	2.7	2.34	3.2	3.2
12	3	2.4	3.1	3.2
13	2.4	2.4	3.3	3.3
14	3	2.3	3.1	3.2

Table C4: Flux recovery after backwashing cycle at frequency 3 hours		
Brushing cycle (hr.)	Backwashing frequency	Backwashing frequency
0		
3	7.2	7.3
3.5	17	17.5
6	9.6	9.8
6.6	15.2	15.5
9	7.5	8.2
9.6	14.4	14.8
12	8.1	8.3
12.5	13.9	14.6
24	6.5	6.9
24.5	13.2	13.6
27	5.1	5.5
27.5	14.1	14.5
30	6.5	6.8
30.5	13.8	14.0
33	7.1	7.2
33.5	14.6	15.0
36	7.3	7.7
36.5	14.2	14.3
48	6.2	6.5
48.5	14.3	14.4
51	7.2	7.4
51.5	13.5	14.0
54	6.8	7.1
54.5	13.8	14.1
57	8.6	9.1
57.5	12.5	12.4
60	4.8	4.5
60.5	12.5	12.4
72	5.4	5.2
72.5	11.8	11.7
75	7.5	7.1
75.5	11.8	11.7

78	6.9	6.7
78.5	12.9	12.8
81	7.1	6.4
81.5	12.8	12.7
84	6.8	6.5
84.5	12.5	12.4
96	6.5	6.8
96.5	12.1	12.2
99	4	4.1
99.5	11.2	11.6
102	6.5	6.6
102.5	10	10.4
105	6.1	6.0
105.5	9.89	9.8
108	5.3	5.2
108.5	10.2	10.1
120	4.2	4.1
120.5	9.8	9.7
123	5.2	5.1
123.5	10.2	10.0
126	6.1	6.0
126.5	9.87	9.8
129	5.1	4.8
129.5	11.3	11.2
132	5.5	5.1
132.5	10.1	10.0
144	3.5	3.3
144.5	9.8	9.9
147	5.1	5.3
147.5	10.3	10.4
150	6.2	6.3
150.5	9.78	10.0
153	6.5	6.6
153.5	10.5	10.6
156	5.5	5.7
156.5	9.8	9.9
168	4.1	4.4

Table C5: Flux recovery after backwashing cycle at frequency 6 hours		
Brushing cycle (hr.)	Backwashing frequency	Backwashing frequency
6	5.6	5.9
6.6	13.2	13.9
12	5.1	4.9
12.5	12.8	12.6
24	4.2	3.9
24.5	12.8	12.7
30	4.3	4.6
30.5	13.2	12.9
36	3.9	4.1
36.5	11.9	12.3
48	3.5	3.7
48.5	12.4	12.6
54	3.5	3.4
54.5	11.8	11.5
60	3.1	3.6
60.5	11.5	10.9
72	2.7	3.1
72.5	10.8	11.02
78	3.1	3.2
78.5	11.8	10.98
84	3.8	3.7
84.5	11.1	11.9
96	2.9	3.1
96.5	10.6	11.3
102	3.5	3.8
102.5	10.6	11.2
108	3.1	3.7
108.5	9.8	10.1
120	2.6	2.9
120.5	10.2	11.2
126	3.4	3.5
126.5	9.87	9.9
132	3	3.5
132.5	10.2	11.2
144	2.7	3.5
144.5	9.8	10
150	2.9	3.2
150.5	8.7	7.89
156	3.1	3.3
156.5	8.4	8.6
168	2.5	2.9

Table C6: Flux recovery after backwashing cycle at frequency 12 hours

Brushing cycle (hr.)	Backwashing frequency	Backwashing frequency
12	4.2	3.9
12.5	10.3	11
24	3.5	4
24.5	11.2	11.9
36	3.1	3.5
36.5	9.8	10
48	3.1	3.4
48.5	10.9	11.6
60	2.5	2.9
60.5	8.7	9.4
72	2.4	2.8
72.5	7.8	8.01
84	2.6	3
84.5	10.2	9.87
96	2.5	3.1
96.5	8.96	8.2
108	2.7	3.01
108.5	10.3	10.5
120	2.3	2.5
120.5	8.3	7.8
132	2.5	3.1
132.5	9.8	10.2
144	2.8	2.5
144.5	10.5	8.7
156	2.7	2.4
156.5	9.7	8.5
168	2.4	2.2

Data for WFMF brushing

Brush was inserted in between the membrane up and down movements of the brush was the form of brushing done and the duration of brushing was 60 seconds per membrane.

Table C7: Effect of brushing every 3 hours

Period (day)	Base fouling curve	Base fouling curve	Brushing	Brushing
1	12.8	13.01	13.5	
2	4.5	4.3	8.36	8.2
3	3.6	3.6	6.8	7.9
4	3.3	3.4	6.54	7.32
5	3.15	36	6.73	5.94
6	2.9	3.01	6.17	6.3
7	2.8	2.8	5.89	5.2
8	2.7	2.78	5.5	4.7
9	2.4	2.5	4.7	4
10	2.3	2.33	4.4	4.3
11	2.25	2.34	4.7	4.1
12	2.35	2.4	4.2	3.5
13	2.2	2.7	3.9	3.3
14	2.3	2.1	4.1	3.6

Table C8: Effect of brushing every 6 hours

Period (day)	Base fouling curve	Base fouling curve	Brushing	Brushing
1	12.4	12.87	12.04	11.87
2	5.4	4.3	7.32	7.58
3	3.8	3.7	5.75	5.67
4	3.5	3	5.17	5.75
5	3.21	2.7	5.2	5.32
6	3.15	3.4	4.82	5.2
7	2.89	3.2	4.37	4.6
8	2.95	3.4	4.2	3.2
9	2.49	2.9	4.1	3.4
10	2.7	2.7	3.9	3.3
11	2.9	3.1	3.8	3.4
12	3.1	2.4	3.5	3.1
13	2.4	2.9	3.6	3.3
14	2.6	2.1	3.4	3

Table C9: Effect of brushing every 12 hours				
Period (day)	Base fouling curve	Base fouling curve	Brushing	Brushing
1	12.92	13.4	13.32	12.94
2	4.7	5.4	5.15	4.54
3	4.1	3.4	4.34	4.24
4	3.8	3	4.54	3.94
5	3.5	3.8	4.24	3.84
6	3.02	2.8	3.94	3.72
7	2.97	2.7	3.54	3.44
8	3.1	3.7	3.3	3.39
9	2.1	2.1	3.4	3.45
10	2	2.01	3.2	3.14
11	2.5	2.34	3.1	2.95
12	2.41	2.4	2.9	3
13	2.34	2.4	3	2.98
14	3.1	2.3	3	2.94

Table C10: Flux recovery after brushing cycle at frequency 3 hours		
Brushing cycle (hr.)	Brushings frequency	Brushing frequency
0		
3	8.4	7
3.5	16.2	16.88
6	7.4	9.18
6.6	15.62	14.88
9	6.8	7.58
9.6	13.2	14.18
12	6.5	7.68
12.5	13.9	13.98
24	5.2	6.23
24.5	13.2	13
27	7.1	4.85
27.5	14.2	13.9
30	8.5	6.19
30.5	13.5	13.38
33	7.1	6.58
33.5	15.2	14.33
36	6.8	7.1
36.5	13.8	13.7

48	6.5	5.9
48.5	14.5	13.8
51	8.6	6.81
51.5	14.1	13.35
54	7.2	6.5
54.5	13.5	13.43
57	6.8	8.43
57.5	13.1	11.76
60	5.1	3.88
60.5	12.8	11.76
72	5.3	4.54
72.5	11.1	11.06
75	6.8	6.48
75.5	11.5	11.06
78	5.9	6.08
78.5	12.5	12.16
81	7.2	5.78
81.5	11.9	12.06
84	6.3	5.84
84.5	12.8	11.76
96	6.8	6.18
96.5	13.1	11.6
99	5.1	3.5
99.5	10.6	10.93
102	4.5	6
102.5	11.2	9.78
105	5.5	5.36
105.5	10.9	9.15
108	5.8	4.56
108.5	11.1	9.46
120	5.1	3.46
120.5	10.1	9.06
123	5.2	4.46
123.5	10.2	9.34
126	5.1	5.36
126.5	11.2	9.13
129	4.2	4.16
129.5	12.4	10.56
132	6	4.48
132.5	11.5	9.36
144	4.52	2.65
144.5	10.5	9.3
147	4.8	4.71

147.5	10.1	9.8
150	5.1	5.7
150.5	11.5	9.41
153	6.2	6
153.5	11.1	10
156	5.1	5.12
156.5	10.2	9.3
168	3.9	3.78

Table C11: Flux recovery after brushing cycle at frequency 6 hours

Brushing cycle (hr.)	Brushing frequency	Brushing frequency
6	5.3	5.2
6.6	12.9	12.8
12	6.1	4.7
12.5	13.2	12.4
24	4.8	3.8
24.5	13	12.4
30	5.1	3.9
30.5	13.8	12.8
36	4.1	3.5
36.5	12.3	11.5
48	4.1	3.1
48.5	11.8	12
54	3.8	3.1
54.5	12	11.4
60	2.9	2.7
60.5	12.1	11.1
72	3.1	2.3
72.5	11.1	10.4
78	3.4	2.7
78.5	12.1	11.4
84	4.2	3.4
84.5	12.1	10.7
96	3.1	2.5
96.5	9.8	10.2
102	4.1	3.1
102.5	9.8	10.2
108	2.9	2.7
108.5	9.7	9.4
120	2.8	2.2
120.5	9.8	9.8
126	3.1	3

126.5	10.2	9.47
132	3.4	2.6
132.5	10.5	9.8
144	3.1	2.3
144.5	9.8	9.4
150	3.5	2.5
150.5	11.2	8.3
156	3.5	2.7
156.5	9.8	8
168	2.9	2.1

Table C12: Flux recovery after brushing cycle at frequency 12 hours

Brushing cycle (hr.)	Brushing frequency	Brushing frequency
12	5.1	4.4
12.5	10.8	10.5
24	4.1	3.7
24.5	11.5	11.4
36	3.5	3.3
36.5	10.1	10
48	3.7	3.3
48.5	11.5	11.1
60	3	2.7
60.5	7.9	8.9
72	3.1	2.6
72.5	9.8	8
84	2.8	2.8
84.5	10.4	10.4
96	2.8	2.7
96.5	9.2	9.16
108	2.5	2.9
108.5	11.3	10.5
120	3.2	2.5
120.5	8.3	8.5
132	3.5	2.7
132.5	9.87	10
144	3.2	3
144.5	11.2	10.7
156	2.5	2.9
156.5	8.9	9.9
168	2.6	2.6

Appendix D Analytical procedure

TURBIDITY

Turbidity checks where done with the HACH portable Turbidimeter Model 2100P. The procedudue followed was as stipulated in the instrument and procedure manual. Turbidity results were used to determine the integrity of the WFMF membrane. The analytical procedure as abstracted for HACH manual CAT.NO. 46500-88 is shown below.

Analytical procedure:

1. Collect a representative sample in a clean container. Fill a sample cell to the line (about 15 mL), taking care to handle the sample cell by the top. Cap the cell.
2. Wipe the cell with a soft, lint-free cloth to remove water spots and fingerprints.
3. Apply a thin film of silicone oil. Wipe with a soft cloth to obtain an even film over the entire surface.
4. Press: **I/O**. The instrument will turn on. Place the instrument on a flat, sturdy surface. Do not hold the instrument while making measurements.
5. Insert the sample cell in the instrument cell compartment so the diamond or orientation mark aligns with the raised orientation mark in front of the cell compartment.
6. Select manual or automatic range selection by pressing the **RANGE** key. The display will show **AUTO RNG** when the instrument is in automatic range selection.
7. Select signal averaging mode by pressing the **SIGNAL AVERAGE** key. The display will show **SIG AVG** when the instrument is using signal averaging. Use signal average mode if the sample causes a noisy signal (display changes constantly).
8. Press: **READ** the display will show **- - - - NTU**, then the turbidity in NTU. Record the turbidity after the lamp symbol turns off.

E-COLI TEST

E-coli tests were carried out on the river samples as well as on the spiked synthetic feed. The E-coli tests were done at the beginning of filtration process. The presence of E-coli and coliform in the feed sample meant that there were micro-organisms in the feed, which had high fouling propensities on the WFMF membrane. Total rejection of the E-coli suggested that the micro-organisms were attached on the membrane surface and this was used to explain the high flux decline in the feeds.

The Colilert Quanti-tray method was used to determine E-coli.

Colilert® is a commercially available enzyme-substrate liquid-broth medium (IDEXX Laboratories, Inc., Westbrook, Maine) that allows the simultaneous detection of total coliforms and *Escherichia coli* (*E. coli*). It is available in the most-probable number (MPN) or the presence/absence (PA) format. The MPN method is facilitated by use of a specially designed disposable incubation tray called the Quanti-Tray®. The PA method can be done using any sterile bottle of the appropriate size. Both methods can be done in the field or laboratory.

THEORY: Two enzyme substrates are included in Colilert—a chromogen that reacts with the enzyme found in total coliforms (galactosidase), and a fluorogen that reacts with an enzyme found in *E. coli* (glucuronidase). After 18 or 24 hours incubation at 35°C (depending on which test kit is used), a total- coliform-positive reaction turns the medium yellow; an *E. coli*-positive reaction causes the medium to fluoresce under a long-wave ultraviolet light (366 nm).

USE: The Colilert method is approved for use with drinking water. It can be used to monitor all types of water, including water that is high in suspended sediment; it can also be used to monitor sediments. The MPN format can be used with all waters; however, it requires the use of a special machine that seals the incubation tray. Quanti-Tray/200 provides counts from 1 to 200 MPN/100 mL. Quanti-Tray/2000 provides counts from 1 to

2,400 MPN/100 mL. Both tests can provide a wider range by diluting the sample 1:10 or 1:100 with sterile distilled or deionized water.

Media Sources:

- The Colilert substrate: Colilert, Cat WP020 (20 tests) or Cat WP200 (200 tests); OR Colilert-18, Cat WP020-18 (20 tests) or Cat WP200-18 (200 tests)
- Incubation trays (Cat WQT100 (Quanti-Tray) or Cat WQT-2K (Quanti-Tray/2000)
- The Quanti-Tray sealer (Cat WQTS2X-115) is purchased from the manufacturer (IDEXX, 800/321-0207).

Standard sample-collection bottles were used. Because the Colilert substrate is already buffered, when preparing dilutions, deionized water was used for the dilutions.

The analytical procedure as abstracted for HACH manual DOC316.53.01449 is shown below.

Analysing samples by most probable number method:

1. Warm up the sealer; this takes about 10 minutes.
2. Prepare lab forms. Fill out appropriate lab forms for each sample and check off the dilutions to prepare, if any.
3. Label the bottom of the incubation tray with sample information. Label the tray with site name, date, and dilution (if any).
4. Prepare sample/reagent mixture.
    a. For undiluted sample:
        i. The system will enumerate between 1 and 2,400 MPN/100 mL for the undiluted sample.
        ii. Combine 100 mL of sample with one packet of Colilert reagent.
        iii. Mix to dissolve reagent by gently inverting the bottle. Do not shake vigorously; this will cause foam.
    b. For dilution:

        i. If you suspect the water to have greater than 2,400 MPN/100 mL, make a 1:10 dilution
        ii. Add 10 mL of the sample to 90 mL of sterile deionized water and combine with Colilert reagent.
        iii. Mix to dissolve reagent by gently inverting the bottle. Do not shake vigorously; this will cause foam.

5. Pour the reagent/sample mixture into the incubation tray. Tap the small wells to release any air and allow foam to settle.
6. Run the tray through the Quanti-Tray sealer.
7. Incubate at 35±0.5°C
    a. Colilert-24 results are definitive at 24–28 hours. In addition, positives observed before 24 hours and negatives observed after 28 hours are also valid
    b. Colilert-18 results are definitive at 18–22 hours. In addition, positives observed before 18 hours and negatives observed after 22 hours are also valid.
8. Count large and small positive wells that
    a. Fluoresce under a long-wave ultraviolet light as *E. coli*
    b. Appear yellow under ambient light as total coliforms
    c. Dim yellow colour and dim or off-colour fluorescence are not counted as positive results.
    d. The large overflow well at the top of the tray is counted as a large well.
9. Refer to the MPN table to obtain results. For a 1:10 dilution, multiply the result in the table by 10 to get MPN/100 mL.
10. Record results on a bench sheet form.

<u>Quality control cultures:</u>

Positive and negative control cultures must be performed once every 20th sample. Control cultures are ordered from the Idexx.

Supplies required

- IDEXX controls: UN3373-WQC Coliform/E. coli for Colilert (IDEXX cat #: 98-29000-00)
- Three Colilert reagent packets
- Three Quanti-Tray/2000
- Three sterile bottles or vessels
- 300 mL of sterile deionized water

One set (3 vials) should be processed per analyst. Use separate small bottles for processing each of the three controls in the set and follow the manufacturer's instructions. Control cultures are used to ensure that field personnel are able to correctly distinguish between positive and negative wells and process the samples correctly. Personnel must obtain results within acceptable limits provided by the manufacturer. Control culture results are not stored in NWIS.

For the Colilert method, three cultures are analyzed:

- *E. coli*—positive for total coliforms and *E. coli*. Wells will be yellow under normal light and fluoresce under long wave UV light.
- *Klebsiella pneumonia*—Positive control for total coliforms, but negative for *E. coli*. Wells are yellow under normal lighting, but do not fluoresce.
- *Pseudomonas aeruginosa*—Negative for total coliforms and *E. coli* but does grow and produces a faint green fluorescence. Wells are counted as negative for yellow color and fluorescence.

CHLORINE TEST

Synthetic feed was prepared with tap water, and domestic sewerage. It was important that the tap water had chlorine levels less than 0.1mg/L so that the experiments were not affected. Presence of chlorine could deactivate bacteria and oxidize the organics coming in with the sewer hence affecting the filtration performance. Source of chlorine was tap water and sodium hypochlorite used for membrane cleaning. To avoid the addition of chlorine tap water was allowed to stay in the sun so that chlorine evaporates and membranes were thoroughly rinsed before experiments were conducted. The synthetic feed and the permeate of the pure water flux (before filtration) were both tested for chlorine to ensure there were no high levels of chlorine in the feed or within the membrane themselves.

Chlorine test was done using HACH Colorimeters, the method used to test was PDP Powder Pillow.

Procedure followed as abstracted from the HACH manual is as follows:

1. Set the instrument to high range (LR), reads chlorine levels from 0.02 – 2 mg/L
2. **Prepare the blank:** Fill a 1-cm/10-mL sample cell to the 5-mL mark with sample. Close the sample cell.
3. Clean the blank sample cell.
4. Insert the blank into the cell holder. Point the triangle mark on the sample cell away from the keypad.
5. Install the instrument cap over the cell holder.
6. Push **ZERO**. The display shows "0.0".
7. Remove the sample cell from the cell holder.
8. **Prepare the sample:** Fill a second 1-cm/10-mL sample cell to the 5-mL mark with sample.
9. Add two 10-mL DPD Free Chlorine Reagent Powder Pillows or two 10-mL DPD Total Chlorine Reagent Powder Pillows to the second sample cell.
10. Close the sample cell. Shake the sample cell for about **20 seconds** to dissolve the reagent. Undissolved power will not affect accuracy. A pink color will show if chlorine is in the sample.

11. Clean the prepared sample cell.
12. **Free chlorine measurement:** Within 1 minute of the reagent addition, insert the prepared sample into the cell holder. Point the triangle mark on the sample cell away from the keypad.
13. Set and start a timer for 3 minutes. A 3-minute reaction time starts.
14. **Total chlorine measurement:** After 3 minutes and within 6 minutes of the reagent addition, insert the prepared sample into the cell holder. Point the triangle mark on the sample cell away from the keypad.
15. Install the instrument cap over the cell holder.
16. Push READ. Results show in mg/L chlorine (Cl2).