CLEANING OF MICRO-AND ULTRAFILTRATION MEMBRANES WITH INFRASONIC BACKPULSING

by

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Declaration

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ABSTRACT

Membrane fouling is universally considered to be one of the most critical problems in the wider application of membranes in filtration separation. Fouling is caused by the deposition of particles not only on the surface of the membrane, but also inside the membrane pores, which reduces permeate flux and leads to a reduction of the efficiency and the longevity of the membrane. The backpulsing cleaning method can be used to remove deposited foulants from the surface of the membrane, without having to shut down the plant. Ultrasonic time-domain reflectometry (UTDR) is a nondestructive technique, used to detect and measure the growth of fouling layer on the membrane surface during microfiltration and ultrafiltration processes.

In this study flat-sheet microfiltration (MF) and ultrafiltration (UF) membranes were fouled during a cross-flow filtration processes using dextrin, yeast or alumina (feed pressure 100 kPa and feed flow rate 0.45 liter/minute), in a flat cell. Infrasound frequency backpulsing, in the permeate space, was used to clean the membranes. Backpulsing was carried out using the permeate water or soap solutions. The peak pressure amplitude of the pulses used to clean the membranes was 140 kPa, the pulsing was applied at a frequency of 6.7 Hz.

The main objectives of this research were: (1) to obtain a fundamental understandimg of how foulants deposit on membrane surfaces and how the foulant deposits can be removed using the backpulsing cleaning technique during MF and UF, (2) to use the ultrasonic measurement technique for monitoring the growth and removal of the fouling layer on the membrane surface and (3) Use scanning electron microscopy (SEM) as a direct measurement technique to analyze the structure the foulant deposits on membrane surfaces before and after cleaning.

Results showed that a flux value of between 55% and 98% of the clean water flux value can be achieved by backpulsing cleaning. UTDR was successfully applied to monitor membrane cleaning and provide information about the growth and removal of fouling layers on the membrane surface.

OPSOMMING

Membraanaanvuiling is wêreldwyd bekend as een van die mees kritieke probleme wat die wyer aanwending van membrane vir skeidingsprosesse benadeel. Aanvuiling word veroorsaak deur die deponering van partikels, nie net op die oppervlak van die membraan nie, maar ook binne-in die membraanporieë, wat die volgende tot gevolg het: 'n afname in vloed deur die membraan, 'n afname in die effektiwiteit van die membraan, en 'n korter membraanleeftyd.

Die teenpulsskoonmaakmetode kan gebruik word om die aanvuilingslaag vanaf die membranoppervlakte te verwyder sonder dat dit nodig is om die membraantoetsapparaat af te skakel. Ultrasoniese-tydsgebied-weerkaatsing (UTW) is 'n nie-vernietigende tegniek wat gebruik kan word om die groei van 'n aanvuilingslaag op 'n membraanoppervlakte tydens mikrofiltrasie (MF) of ultrafiltrasie (UF) te identifiseer en te meet.

In hierdie studie is plat-vel MF en UF membrane bevuil gedurende 'n kruisvloeifiltrasieproses deur gebruik to maak van dekstraan, gis of alumina, in 'n plat sel. Infraklank-frekwensie-teenpols, in die permeaatgebied, is gebruik om die membrane skoon te maak. Hiervoor is die proseswater of 'n seepoplossing gebruik. Die maksimum drukamplitude van die pulse wat gebruik is was 140 kPa, en die puls was aangewend teen 'n frekwensie van 6.7 Hz.

Die hoofdoelwite van hierdie studie was die volgende: (1) om inligting in te win oor hoe aanvuilingsmateriale op membraanoppervlaktes gedeponeer word tydens MF en UF en hoe hulle verwyder kan word deur gebruik te maak van die teenpulsskoonmaaktegniek; (2) om van die teenpulsskoonmaaktegniek gebruik te maak om die groei van die bevuilingslaag asook die verwydering daarvan op die membraanoppervlakte te monitor; en (3) om van skandeerelektronmikroskopie (SEM) as 'n direkte analitiesetegniekgebruik te maak om die struktuur van die aanvuilingsmateriaal voor en na die die skoonmaakproses te analiseer.

Deur gebruik te maak van teenpulsskoonmaak kon die membraanvloed tot tussen 55–98% van die oorspronklike suiwerwatervloed verbeter word. Sodoende is ultrasoniese-tydsgebied-weerkaatsing suksesvol gebruik om die skoonmaak van membrane te monitor asook om inligting in te win i.v.m. die groei en verwydering van die aanvuilingslae op die membraanoppervlaktes.

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LIST OF ABBREVIATIONS

BP	Backpulsing
BSA	Bovine serum albumin
CA	Cellulose acetate
CML	Carboxylate modified latex
CP	Concentration polarization
EDTA	Ethylene diamine tetra-acetic acid
FP	Feed pressure
LY	Lysozyme
MF	Microfiltration
MWCO	Molecular weight cut-off
NF	Nanofiltration
OV	Ovalbumin
PA	Polyamide
PBP	Peak of backpulse pressure
PC	Polycarbonate
PE	Polyethylene
PES	Poly(ether sulfone)
PI	Polyimide
PS	Polysulfone
PVDF	Polyvinylidene difluoride
RO	Reverse osmosis
SEM	Scanning electron microscopy
SLS	Sodium lauryl sulfate
TMP	Transmembrane pressure
TFC	Thin-film composite
UF	Ultrafiltration
UTDR	Ultrasonic time-domain reflectometry
XRD	X-ray diffraction
XRF	X-ray fluorescence

CHAPTER 1

1.1 INTRODUCTION

Membrane filtration was not considered a technically important separation process until about 25 years ago. Nowadays, membrane filtration technology can be found in a wide range of applications in many industrial fields, for example, in the food and beverage, diary, biotechnology, metallurgy, pulp and paper, textile, pharmaceutical, and chemical industries, and in water treatment (sea water and brackish water desalination and also microfiltration and ultrafiltration purification of non-saline surface water) for domestic and industrial water supply.[1]

Unfortunately membrane fouling is a major problem in membrane filtration technology. Fouling is often caused by the adsorption of solutes not only on the surface of the membrane, but also inside the membrane pores, which reduces both permeate flux and membrane selectivity, and leads to a reduced life time and efficiency of the membranes.[2]

Various techniques exist to reduce membrane fouling, for example: chemical cleaning, backpulsing, physical brushing, modification of membrane chemistry, feed particle addition, feed pretreatment and hydrodynamic techniques (such as turbulent flow, air sparging, and adding inserts), increasing surface roughness to introduce flow instability, periodic pulsation or using curved channels.[3] Many of these methods can effectively reduce membrane fouling, but they seem not to be sufficiently efficient for the removal of deposited foulants.[3] Backpulsing is a cleaning technique that has been shown to remove deposited foulants from the surface of the membrane.[3]

Backpulsing involves reversing the permeate flow through the membrane for very short periods of time.[3] The reverse flow can provide in situ cleaning by removing some of the foulants from the surface of the membrane.

Several groups, using various foulants, have observed flux enhancement by using back pulsing.[3-8]

Many techniques have been used to analyze the fouling deposits on the membrane surfaces, including scanning electron microscopy (SEM), X-ray fluorescence (XRF), and X-ray diffraction (XRD).[9] These techniques supply some information on the fouling mechanism, but

provide little information about the dynamic growth of the fouling layer on the membrane surface. The ultrasonic technique, as used in this study is a nondestructive and noninvasive technique that can be used for in situ monitoring of the growth of the fouling layer on the membrane surface.[9] This technique has been investigated by several research groups, who found that it provides good information about the growth of the fouling layer on the membrane surface and can be also used to monitor the efficiency of membrane cleaning methods.[9-12]

1.2 MEMBRANE TECHNOLOGY

Membrane separation was first observed in 1748 by the French scientist, Abbe Nollet. He observed that if he stored salt brine inside a pig's bladder and placed it in pure water then the water would pass through the bladder.[13,14] The general definition of a membrane can be a semipermeable membrane which is a thin barrier between two fluids which limits the movement of one or more components of one or both fluids across the barrier.[13]

The operation of a membrane will largely depend on its structure and pore size. Synthetic membranes can be classified into two types according to the structure of the membrane, namely symmetric and asymmetric. Asymmetric membranes have a non-uniform structure comprising an active top layer supported by a non-active porous support.[15] The development of asymmetric membranes in the 1960s led to a breakthrough in the use of membrane separation technology for industrial applications.[1]

There are several membrane separation processes; including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). Reverse osmosis emerged as a useful separation process after successes were achieved using cellulose acetate (CA) membranes in the late 1950s by Reid and Breton, and in the 1960s by Sourirajan and Loeb. Sourirajan and Loeb are credited with making the first high performance RO membranes from CA. Commercially RO membranes were produced in the late 1960s by Gulf General Atomics, these membranes were Sourirajan - Loeb CA membranes in a spiral wound module.[2] The first commercially successful industrial UF system, equipped using the tubular membrane configuration, was produced by Abcor (now a division of Koch Industries, USA) in 1969. it was used to recover electrocoat paint from automobile paint shop rinse water.[16] Membranes are now used on commercial scale for many different applications, for example: the production of potable water from the sea by RO, fractionation of macromolecular solutions in

the food and drug industries by UF, and the purification of drinking water and treatment of industrial wastewater by MF.[17]

The advantages of membrane technology can be summarized as follows.[1]

- The consumption of energy is generally low.
- Separation can be achieved continuously.
- Membrane properties are changeable and can be adjusted.
- Membrane processes can be used in tandem with other separation processes.
- Separation can be achieved under mild conditions.

The disadvantages of membrane technology include, however:[1]

- Concentration polarization and membrane fouling.
- Short membrane lifetime.
- Low selectivity or flux.

1.3 OBJECTIVES

The main objectives of this research were: (1) to ascertain how foulants deposit on membrane surfaces and how the foulant deposits can be removed using the backpulsing cleaning technique during MF and UF. (In the experimental work suspensions or solutions of washed yeast, alumina powder or dextrin were to be used and experiments carried out using flat-sheet MF membranes and UF membranes) and (2) to use the ultrasonic measurement technique for monitoring the growth of the fouling layer on the membrane surface. The following specific objectives were also undertaken:

- Determine which pulse amplitudes from the diaphragm pulsating pump, give the best results for permeate flux values.
- Determine the highest flux values that can be obtained, as a percent of the clean water value, for various combinations of foulants, using alumina, yeast and dextrin.
- Investigate the possibility of using the ultrasonic measurement technique to understand the mechanism of fouling which occurs during MF and UF experiments.
- Use scanning electron microscopy (SEM) as a direct measurement technique to analyze the structure the foulant deposits on membrane surfaces before and after cleaning.

- Determine the efficiency of the backpulsing cleaning method using the ultrasonic measurement technique.

CHAPTER 2

HISTORICAL AND THEORETICAL BACKGROUND

2.1 PRESSURE-DRIVEN MEMBRANE PROCESSES

The heart of every membrane separation process is the membrane, which can be considered as a permselective barrier between two phases. There are several membrane processes such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO), which differ mainly on the basis of their separation mechanism and the size of particles to be separated. The solvent and different solute molecules permeate through the membrane due to the applied pressure, which is considered the driving force of pressure-driven membrane processes, on the other hand, other molecules or particles are rejected to different extents depending on the structure of the membrane[1]. Table 2.1 tabulates the different membrane processes and separation mechanisms.[18]

Table 2.1 Summary of common pressure-driven membrane processes, membrane materials and separation mechanisms[18]

Membrane process	Membrane material	Separation mechanisms
Microfiltration	Polypropylene (PP) Polycarbonate (PC) Ceramics (CC) Nylon Polyamides (PA)/polyimide (PI)	Sieving
Ultrafiltration	Polysulfone (PS) Cellulose acetate (CA) Polyvinylidene fluoride (PVDF)	Sieving
Nanofiltration	Polyvinylalcohol Polyamide (TFC)	Solution diffusion
Reverse osmosis	Cellulose acetate Polyamides (PA) / Nylon	Solution diffusion

It is also possible to discern between the processes in terms of the membrane structure. In the case of MF the thickness of the membrane can extend from 10 μm to more than 150 μm. Ultrafiltration, nanofiltration and reverse osmosis membranes have relatively dense, thin top layers (thickness 0.1 – 1.0 μm) supported by a porous substructures (thickness 50 – 150 μm). Table 2. 2. gives the comparison of the pressure-driven membrane processes.[1]

Table 2.2 Comparison of the common pressure-driven membrane processes[1]

Feature / process	Microfiltration	Ultrafiltration	Nanofiltration/reverse osmosis
Species separation	Separation of suspended solids (e.g. colloidal particles)	Separation of macromolecules (bacteria, yeasts)	Separation of low MW solutes (salts, glucose, lactose, micropollutants). Rejection of divalent ions is higher than of monovalent ions.[13]
Applied pressure	Low (< 2 bar)	Low (1 – 10 bar)	High (10 – 60 bar)
Membrane structure	Symmetric structure	Asymmetric structure	Asymmetric structure
Thickness of actual separating layer	10 – 150 μm	0.1 – 1.0 μm	0.1 – 1.0 μm
Basis of separation	Separation based on particle size	Separation based on particle size	Separation based on differences in solubility and diffusivity

2.1.1 MICROFILTRATION

Microfiltration (MF) is a pressure driven membrane process that uses porous membranes to separate suspended particles, with diameters ranging from 0.1 to 10 μm. The range of the pore sizes of MF membranes is from 0.05 to 10 μm.[16] MF is applied in two types of filtration processes, cross-flow filtration and dead-end filtration, related to the hydrodynamics of the feed flow. Cross-flow filtration can be used to separate suspensions with high solids content, whereas dead-end filtration can be used for low solids content.[19] A schematic of these processes is shown in Figure 2.1.

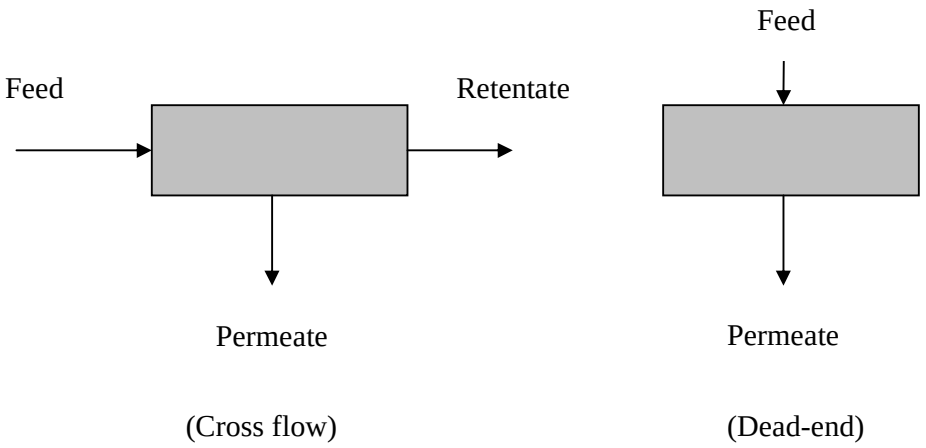


Figure2.1 Schematic representation of cross-flow and dead-end filtration.

MF has many applications, for example, it can be used as a pretreatment for RO plants,[20-22] for separating emulsions (e.g. oil-polluted industrial effluents),[23] for concentrating and washing different colloidal suspensions (pigments, metal hydroxides, grinding effluents)[24,25], in industrial applications, and for wastewater treatment.[26]

MF membranes are generally (polymeric membranes),[1] for example:

- polyamide (PA)
- polysulfone (PS)/poly(ether sulfone) (PES)
- cellulose esters
- polycarbonate (PC)
- polyimide (PI)
- nylon

2.1.2 ULTRAFILTRATION (UF)

Ultrafiltration (UF) is a pressure-driven membrane process, with the separation capability between capabilities of MF and NF. The pore sizes of UF membranes range from 0.05 μm to 0.001 μm. UF membranes can be used to separate dissolved macromolecules and colloids from solutions in the molecule size range 0.001 – 0.02 μm. Solvent and salts of low molecular weight permeate through the membranes, while the larger molecules are rejected. This mechanism is illustrated in Figure 2.2.[3,6]

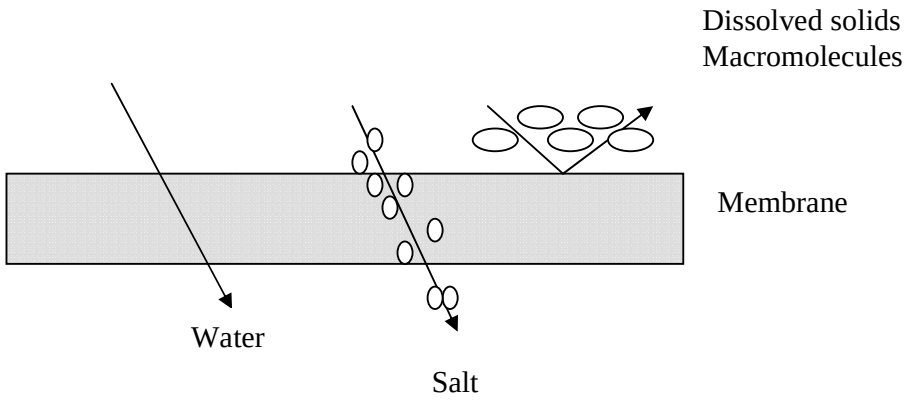


Figure 2.2 Schematic of separation by UF.

Commonly the best method for classifying UF membrane performance is by the molecular weight cutoff (whereas, MF membrane performance is classified by SEM, because the MF pore size can be observed by SEM analysis). In UF membranes the pores are too small for detection by SEM. Furthermore, the pores usually close when samples are dried for the SEM analysis, this makes observation of the surface by SEM difficult.[27]

In general UF is used to separate macromolecular solutes and colloidal material from solvents. UF has many industrial applications, e.g. in the dairy industry, food industry, chemical industry, paper industry, and pharmaceutical industry, and moreover it is used in water treatment, electro-paint and metallurgy (oil and water emulsions).[28-31] Most UF

membranes are polymeric; the membranes are made from different polymer materials,[32] for example:

- polysulfone
- polyimide
- cellulose acetate
- polyethersulphone
- aliphatic polyamides

2.1.3 NANOFILTRATION (NF)

Nanofiltration (NF) is the third pressure driven membrane process and its application lies between those separations using UF membranes and RO membranes. There are similarities between the NF and RO processes. In general NF systems operate at pressures lower than those used for RO. The molecular weight cutoff method can be used to classify NF membranes characteristics. NF membranes can be made by interfacial polymerization on a porous substrate of PS or PES.[33]

NF membranes can be used to separate ions from solutes, removal of hardness, and removal of colour.[34] These membranes also have applications in the dairy industry for cheese desalting,[35] and in water treatment.[36-38] The features of NF are given in Table 2.3.

Table 2.3 Features of nanofiltration[1]

Membrane type	Thin-film composite (TFC)
Pore size	< 2 nm
Pressure driving force	10 – 25 bar
Thickness	sublayer ≈ 150 μm; toplayer ≈ 1 μm
Membrane material	polyamide
Main applications	water softening waste water treatment removal of micropollutants

2.1.4 REVERSE OSMOSIS (RO)

Reverse osmosis (RO) is a pressure driven membrane process used to separate all solute species (organic and inorganic) from solution, and to separate the ionic solutes and macromolecules from solution. The separation mechanism of macromolecules and species is given in Figure 2.3. RO is the opposite of the natural phenomenon of osmosis. When a semi-permeable membrane is used to separate a concentrated solution, RO will take place after applying a pressure greater than the osmotic pressure of the concentrated solution.

If the applied pressure is above the solution's natural osmotic pressure, the solvent will flow through the semipermeable membrane to create a more concentrated solution on the side where pressure is applied and a dilute solution on the other side. If the applied pressure is the same as the solution's natural osmotic pressure no flow will take place. If the applied pressure is lower than the solution's natural osmotic pressure, opposite flow will happen.[2]

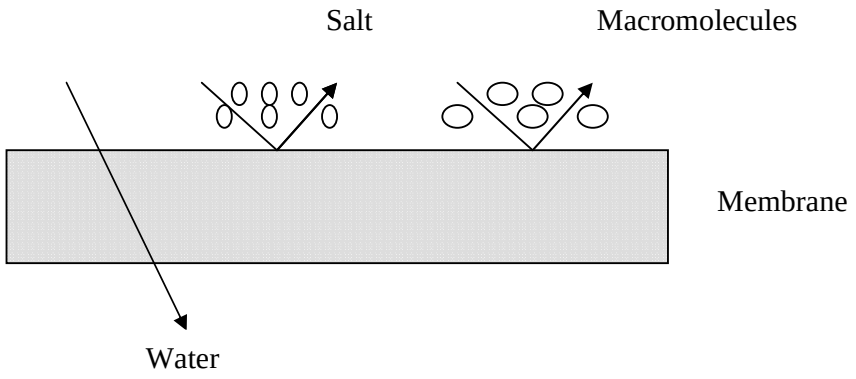


Figure 2.3 Schematic of separation by reverse osmosis.[15]

Reverse osmosis membranes are either composite or asymmetric and generally have a thickness < 1 μm for the dense top layer supported by a 50 – 150 μm thick porous layer. The

substructure of these membranes is made from materials such as cellulose triacetate, aromatic polyamide, and poly(ether urea) or by using interfacial polymerization.[15]

There are many applications of reverse osmosis separation in different industries, such as the food and beverage industry, pharmaceutical industry, and production of pure water for different industries such as boiler feed and electronics applications. The most important application in the purification of water is the desalination of brackish and sea water to produce drinking water.[2] The features of RO are given in Table 2.4.

Table 2.4 Features of reverse osmosis[1]

Membranes type	Asymmetric or composite
Pore size	< 2 nm
Pressure driving force	brackish water 10 – 25 bar sea water 40 – 80 bar
Thickness	sublayer ≈ 150 μm; toplayer ≤1 μm
Membrane material	aromatic polyamides, cellulose triacetate, polyamide and polyether(urea)
Main applications	desalination of brackish and sea water food and dairy industry production of ultrapure water (electronics industry)

2.2 MEMBRANE MODULES

In membrane separation technology there are several different module designs available. There are also some different modules such as plate-and-frame modules, tubular modules, spiral-wound modules and hollow-fiber modules.[15]

There are two types of membrane configuration, used in several possible modules, namely flat-sheet and tubular. Plate-and-frame and spiral-wound modules are based on the flat-sheet membranes configuration, while tubular and hollow fiber modules are based on the tubular membrane configuration.[1]

2.2.1 PLATE-AND-FRAME MODULE

Typically, plate-and-frame membranes are available for MF, UF, NF and RO processes. These modules are one of the earliest modules of membrane technology. The earliest design of a plate-and-frame was proposed by Stern,[36] to recover helium from natural gas.[16] These modules are formed by a number of layers of membranes, feed spacer plates and permeate spacers. Generally the feed spacer plate is made from an appropriate plastic and contains channels which guides the feed solution to flow from the inlet to the plate, to the outlet.[15]

There are several different designs of plate-and-frame modules, such as the design that is illustrated in Figure 2.4. This design has a membrane plated cylinder inside a tubular pressure vessel. The advantages and disadvantages of the frame-and-plate module are listed below.[2]

Advantages

- open flow channels
- easy disassembly for cleaning and membrane replacement
- low tendency to foul
- numerous different membrane types can be used

Disadvantages

- expensive
- low membrane surface area to volume ratio
- possibility of leaks between leaves.

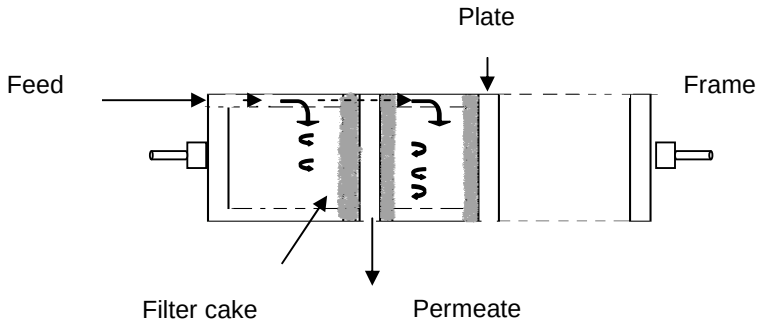


Figure 2.4 Schematic of a plate-and-frame membrane module.[2]

2.2.2 SPIRAL-WOUND MODULE

The spiral wound module utilizes flat-sheet membranes. The membrane module (Figure 2.5) is considered as an envelope. It consists of two flat sheet membranes and a highly porous support material that is placed between the two flat membranes (permeate spacer), which are attached together along three edges with suitable adhesive glue. The fourth edge of the envelope is connected to the permeate tube. A number of spiral-wound modules are connected in series around the collecting tube, which is called the element (see Fig 2.5).[19, 37] The advantages and disadvantages of the spiral-wound module are listed below.[2]

Figure 2.5 Photograph of a spiral-wound membrane element.

Advantages

- easy to clean
- easy field replacement
- good resistance to fouling
- can be made from several different membrane materials
- available from different manufacturers

Disadvantages

- moderate membrane surface area to volume ratio
- concentration polarization is prone to occur
- difficult to obtain high recoveries in small systems

2.2.3 TUBULAR MODULE

Tubular membrane modules generally consist of a membrane formed on the inside of a pressure resistant tube, which is between 5 and 25 mm in diameter. Typically these tubes are made from non-woven fabric, for example, polyester, polypropylene, polyethylene, or fiber-reinforced epoxy tubes.[38] There are numerous options for the module design, such as many smaller tubes nesting inside a large tube. The tubular membrane system consists of a large number of tubes connected together in parallel.[15, 16] Figure 2.6 shows an example of single tubular membrane module. The advantages and disadvantages of the tubular membrane module are listed below.[2]

Advantages

- easy to clean
- low tendency to blockage
- high flow velocities
- membranes can be removed and renewed
- this module can be used at high pressures

Disadvantages

- expensive
- very low membrane surface area
- made from limited materials

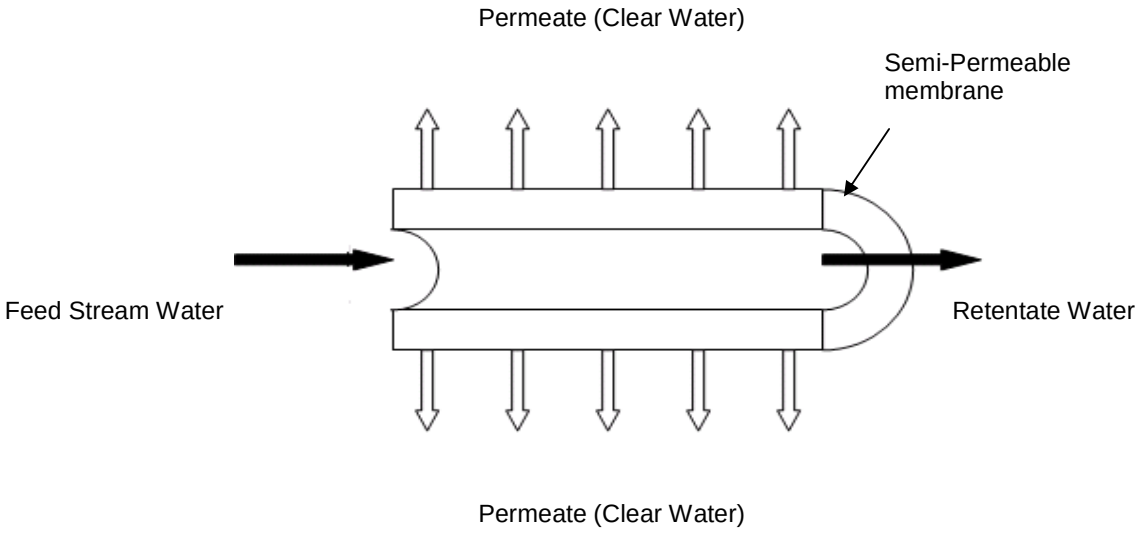


Figure 2.6 Schematic of a tubular membrane module.

2.2.4 HOLLOW-FIBER MODULE

This membrane module consists of hollow-fibers, with diameters usually less than 1 mm, fixed in a vessel as shown in Figure 2.7. The difference between a hollow-fiber module and all the other modules is that there is no supporting layer for the membrane. The actual membrane might be on the inside surface of the fiber tube, the outside surface, or on both surfaces.[15,39,40] Figure 2.7 shows an example of a hollow-fiber membrane module. The advantages and disadvantages of the hollow-fiber membrane module are listed below.[2]

Advantages

- the surface area to volume ratio of the membrane is high
- high recovery
- changing the membrane bundles in the field is easy
- easy to spot problems

Disadvantages

- sensitive to fouling
- minimal choice of membrane materials
- limited number of manufacturers produce this type of module

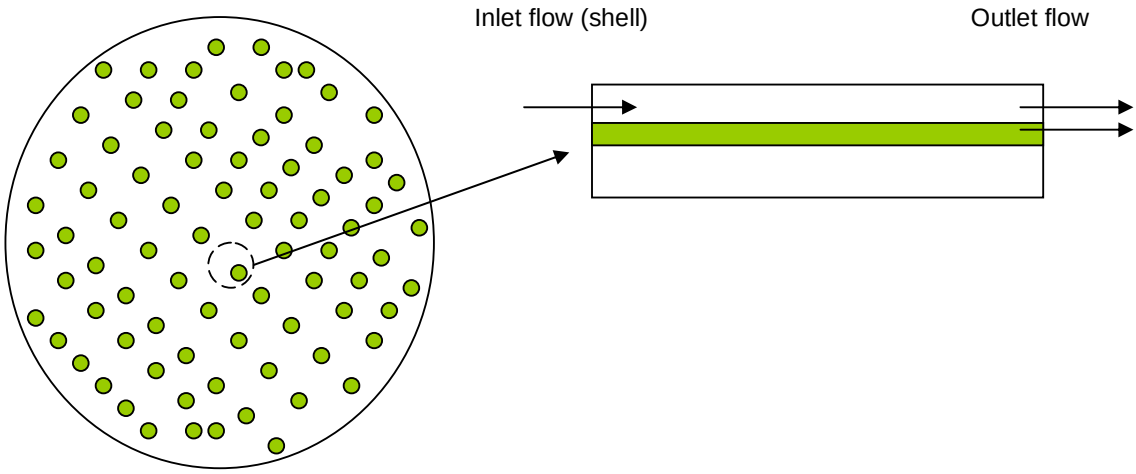


Figure 2.7 Schematic of a hollow-fiber membrane module.[39]

2.3 CONCENTRATION POLARIZATION AND MEMBRANE FOULING

2.3.1 CONCENTRATION POLARIZATION (CP)

In pressure-driven membrane filtration processes such as MF, UF and RO selected components in the solution are rejected by the membrane. In the case of a solution consisting of a solvent and solute, when a suitable pressure (driving force) is applied to the feed solution the solute is partially rejected by the membrane, while the solvent permeates through the membrane. The rejected solutes will accumulate at the membrane surface and their concentration will slowly increase. For this reason the rejected solutes will accumulate on the membrane surface. This phenomenon is called CP.[1] CP is considered to be the main reason for flux decline during the early stages of a membrane separation process.[41] CP has several negative effects:[42]

- The high concentration of solute on the membrane surface can cause changes in the composition of membrane material (due to pore blockage or precipitation within the membrane).
- There is an increase in the hydrostatic resistance due to formation of a gel or cake layer on the membrane surface.

- The driving force for the filtration decreases because of an increase in chemical potential.
- The most important in RO is increased scaling potential on the surface

The effect of CP is very severe in MF and UF, both because the fluxes are high and the mass-transfer coefficients are low, due to the low diffusion coefficients of macromolecular solutes. On the other hand, in RO the flux is lower and the mass-transfer coefficient is higher, and because of this CP has a less severe effect on the RO process.[1,27] Table 2.5 summarizes the effects of CP on the different membrane processes.

There are several methods to reduce CP, for example: increasing the flow velocity, using turbulence promoters, using a pulsating flow to break the boundary layer, and increasing the feed temperature.[27]

Table 2.5 Effects of concentration polarization on the various pressure-driven membrane processes

Membrane process	**Effect**	**Result**
Microfiltration	strong	J large / K small
Ultrafiltration	strong	J large / K small
Nanofiltration	moderate	K large
Reverse osmosis	moderate	K large

J: flux; K: mass transfer coefficient.

A number of experimental and mathematical studies have been done to obtain a better understanding of the CP phenomenon.[41] Gowman and Ethier[43] used an automated laser-based refractometric technique during dead-end filtration of a biopolymer solution for measuring the solute CP gradients, but they found that the data did not agree with theory. The same technique was used by Pope et al.[44] during cross-flow filtration of oil-water emulsions, for measuring the CP layer thickness. The images showed that the technique offered visualization of the CP layer and a non-invasive measurement of its thickness. Electrical conductivity microprobes were used by Liu and Williams[37] to measure the salt CP in an unstirred batch cell, and they observed that the data were in agreement with theory.

2.3.2 MEMBRANE FOULING

The decline in permeate flux with time is considered one of the most serious problems in pressure-driven membrane processes. The flux decline is the result of membrane fouling. Membrane fouling can be defined as the irreversible deposition of retained particles, macromolecules, colloids, salts, etc on or in the membrane. The main modes of membrane fouling include adsorption, chemical interaction, cake formation and pore blocking.[15, 27, 45, 46]

Membrane fouling affects the performance of a membrane either by the deposition of a layer onto the membrane surface or by blockage of the pores. The different modes of blockage of the pores are the following:[15,47]

- complete pore blocking (the pore entrance is tightly closed)
- pore bridging (partial pore blocking)
- internal pore blocking (the material is adsorbed by or trapped on the pore wall of the membrane).

Membrane fouling can be appreciable in MF and UF membranes, which are classified as porous membranes, while membrane fouling can largely be avoided in NF and RO membranes which are classified as dense membranes. Membrane fouling is very complex, and is therefore difficult to describe theoretically. There are physical and chemical parameters that affect fouling, for example, temperature, pH, and solution concentration .[1] Much literature is available on membrane fouling.[46, 48-51]

Because membrane fouling is a particularly serious problem in the application of membrane technology for protein purification, many studies concerning protein fouling have been carried out. Guell and Davis[52] studied the flux decline during microfiltration of protein mixtures of bovine serum albumin (BSA), lysozyme (LY) and ovalbumin (OV) through polysulfone and polycarbonate membranes, and found that the greatest decline was found for the mixtures containing OV. General conclusions indicated that the resistance of MF membranes increases because proteins adsorb under static conditions.[53-55]

There are three types of foulants that can be distinguished:[1]

- organic precipitates (biological substances, macromolecules, etc.)
- inorganic precipitates (metal hydroxides, calcium salts, etc.)
- particulate matter.

Organic precipitates

Organic fouling occurs due to deposition of proteins, dissolved macromolecules and other organic substances on the membrane surface. When the feed solution flows over the membrane surface, the solute molecules can adsorb onto the membrane surface due to physico-chemical interaction. Membrane fouling by proteins takes place in two steps:[56]

- protein adsorption/deposition
- cake formation on the membrane surface.

Inorganic precipitates

Precipitates can be formed by colloidal that critically foul RO membranes in particular. During separation, the precipitate formed is too porous to be harmful, except when there is a change in pH during filtration, during cleaning, or when precipitation occurs inside the membrane pores.[13]

Particulate matter

Examination of the fouling of membranes by particulate matter suggests that the dynamics of flux decline is related to the degree of cake formation on the membrane surface.[13]

2.3.2.1 Mathematical models of concentration polarization and fouling

Fouling is very specific to the particular application of membrane technology and, because of its complex nature, it is difficult to describe it in general terms. Several mathematical models have been developed to try to describe or understand concentration polarization and the fouling phenomenon, for example; cake-filtration model[57, 58]osmotic pressure model[59, 60] and gel polarization model.[61, 62]

Cake-filtration model

In this model the solute is considered to form a deposit of particles on the membrane surface with constant concentration (Figure 2.8). The cake-filtration model is commonly used to determine a fouling index, the flux (Jv) can be described by.[27]

$$Jv = \frac{\Delta P}{\eta(Rm + Rc)} \qquad (2.1)$$

Where ΔP is the applied pressure, Rm is the membrane resistance; Rc is the total cake layer resistance; η is the solution viscosity.

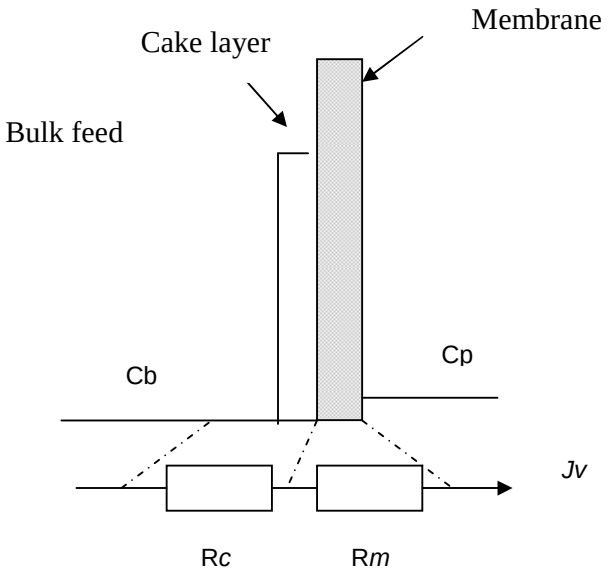


Figure 2.8 Schematic representation of the cake-filtration model.[27]

Cb : concentration of the solute in the feed side

Cp : concentration of the solute in the permeate side

Rc : total cake layer resistance

Rm : membrane resistance

2.3.2.2 Methods to reduce fouling

The reduction of fouling is very specific to the process and depends very much on the application. The methods used to enhance the performance of the membrane can be classified in the following three categories: [27]

- pretreatment of the feed solution
- changing the membrane properties
- changing the process conditions

Pretreatment of the feed solution

Typical methods of feed solution pretreatment include heat treatment, addition of a complexing agent (e.g. EDTA), pH adjustment to prevent scaling, chlorination, chemical clarification, addition of activated carbon, pre-microfiltration and pre-ultrafiltration. In the case of feed solution concentrating proteins pH adjustment is very important.[27]

Changing membrane properties

Changing the membrane properties is one method that can be used to reduce fouling. Generally fouling with MF and UF membranes is more critical than with NF and RO membranes. Chemical modification of membranes, for example, sulfonation of polysulfone and blending a hydrophobic polymer with a hydrophilic, can reduce fouling.[1,27]

Changing process conditions

The most important factor in reducing CP and fouling is increasing the mass transfer coefficient. Increasing the flow velocity and using lower-flux membranes will increase mass transfer. Furthermore, the use of different kinds of turbulence promoters can also reduce fouling.[1] Fouling can be controlled by operating at the critical flux, where flux versus back diffusion in the CP layer counteract each other. [63,64]

2.3.2.3 Membrane cleaning

There are commonly three cleaning methods to reduce fouling: hydraulic cleaning, mechanical cleaning and chemical cleaning. The choice of the cleaning method depends on the module configuration and the chemical resistance of the membrane. Some methods are:

Hydraulic cleaning

There are a number of hydraulic cleaning methods, such as back-flushing (used only with MF and UF membranes), back-shock (back-flushing for only a fraction of a second), and backpulsing.[27] Backpulsing is discussed in the next section (2.4.1).

Mechanical cleaning

This method has only been applied to tubular membrane systems, using oversized sponge balls.[1]

Chemical cleaning

The most important cleaning method for removing fouling is chemical cleaning. Many chemical agents are available for removing or dissolving the deposits from a membrane. Chemical cleaning involves the use of chemicals to react with the deposits and other foulants that affect the flux rate and permeate water quality. The concentration of the cleaning agent and the duration of cleaning are very important in membrane cleaning and can also affect the chemical resistance of the membrane. Some important chemicals used to clean the membrane are[1];

- acids (sulfamic acid, oxalic acid and citric acid)
- alkalis (phosphates, carbonates and hydroxides)
- complexing agents (ethylenediamine tetra-acetic acid (EDTA), polyacrylates and sodium hexametaphosphate)
- enzymes (proteases, amylases and glucanases).

2.4 REDUCTION OF MEMBRANE FOULING BY THE BACKPULSING TECHNIQUE

2.4.1 DESCRIPTION OF BACKPULSING (BP)

The common cleaning methods (section 2.3.2.2) can reduce membrane fouling, but they have a minimal effect on deposited foulants. Cleaning by backpulsing (BP) is much better than these methods as BP can remove the deposited foulants from the surface of a membrane.[3, 65-67]

Today (BP), or transmembrane pressure (TMP) pulsing, is considered an effective method to reduce membrane fouling and to improve the efficiency of membrane separation processes.[66]

The BP process is illustrated in Figure 2.9.[4] during the forward filtration, the applied pressure on the feed side is much greater than the pressure in the permeate side, and hence the feed liquid is forced to flow to the permeate side. In reverse filtration (backpulsing), the pressure on the permeate side is higher that the pressure on the feed side (reversed TMP), and hence the permeate liquid is forced back through the membrane to the feed side and dislodge the deposit of rejected foulants on or from inside the membrane.

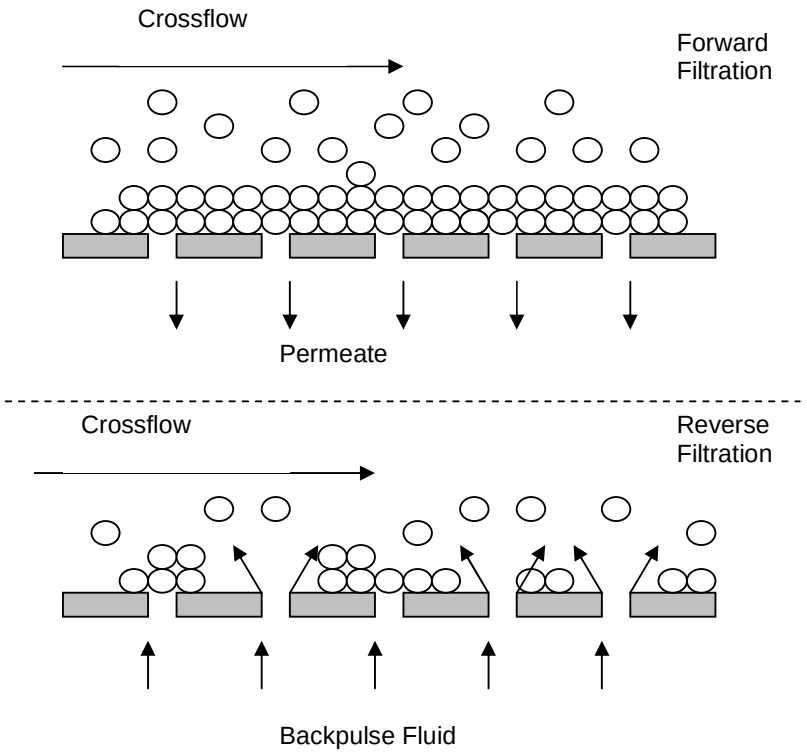


Figure 2.9 Schematic of the backpulsing process during forward and reverse cross-flow filtration.[4]

This reverse flow lifts away from the membrane a portion of the deposited foulants, which are then removed from the membrane module by the cross-flow.[4, 5] There are several factors affecting the backpulsing cleaning method: backpulse duration (the period of time that the filtration system operates under negative transmembrane pressure), pulse amplitude (the absolute value of average transmembrane pressure during backpulsing), and backpulse interval (the time duration between two consecutive pulses).[5]

Typically, the backpulsing method is a variation of the backflushing or backwashing method. The principle difference between backpulsing and backflushing is the negative TMP force and speed used to remove deposited foulants from the membrane surface. In backflushing the reverse TMP occurs for 5 – 30 s every 30 min to many hours, in backpulsing, the reverse pressure pulses are applied for very short periods of time (typically less than 1 s), and at high frequency (typically 0.1 – 2 Hz). [6, 68]

Infrasonic frequency pulsing is now considered as a new technique to reduce fouling with frequencies in the order of 1 – 10 Hz and pressure pulses that are applied on the permeate side to remove the deposition of foulants on the feed, this is accomplished by vibrating the membrane. Consequently, one cycle of infrasonic frequency pulsing might be divided into three stages [69, 70]:

Stage 1: Formation of the cake causing the permeate flux to decrease
Stage 2: Application of an infrasonic pulse on the permeate side which causes the membrane to vibrate.
Stage 3: Reverse movement of the membrane during pulse decay, which will lead to some of the foulant cake to becoming detached from the membrane surface.

The backpulsing method of cleaning has been studied by many groups. Rodgers and Sparks[71-73]and Wilharm and Rodgers[67] used backpulsing during UF experiments, including UF of dilute protein solutions (bovine serum albumin) as the foulant, and flat-sheet polymeric membranes. They found that for laminar cross-flow the flux values after backpulsing increased to 100 times that of the unpulsed flux, but for turbulent cross-flow, backpulsing had little effect on the permeate flux. They concluded that the reason for the flux increase was the concentration polarization disruption by the motion of the membrane and not by backflow through the membrane.

Ramirez and Davis[6] used backpulsing with cross-flow MF for water treatment, using two types of membranes: a tubular ceramic membrane and a hollow fiber cartridge membrane. The experiments were performed with suspensions of bentonite clay in water and with dilute oil-in-water. They found that the permeate flux in the pulsed clay suspension experiments increased more than 10-fold, whereas in the oil-in-water experiments the flux increased up to 25 times.

Ma et al.[7] used backpulsing with MF of carboxylate modified latex (CML), using polypropylene membranes, and recorded an approximately two-fold permeate flux enhancement over 1 hour of filtration when using backpulsing. Wenton and coworkers[74, 75] used backpulsing to clean hollow-fiber membranes fouled by beer, rennet and cellulose, and found that use of backpulsing resulted in stable permeate fluxes at low crossflow velocity and TMP.

Redkar and Davis[8] used backpulsing during MF of washed yeast cell suspensions, using flat sheet CA membranes, and found that the permeate flux increased 10-fold. Redkar et al. [76] used backpulsing with MF of yeast suspended in deionized water and obtained permeate fluxes that were up to 85% of that of the clean membrane flux. Parnham and Davis[77] used backpulsing to recover the protein from a bacterial cell lysat. They found that the net flux increased when the forward and backpulse pressure were increased.

Sondhi et al.[5] investigated the use of backpulsing as an effective method for decreasing fouling during crossflow filtration for synthetic wastewater suspensions containing chromium as the main constituent, with ceramic membranes. They concluded that backpulsing was an effective method to reduce the fouling, as the permeate flux increased 5-fold with backpulsing.

Czekaj et al.[70, 78] used infrasonic frequency pulsing during MF of suspensions of 0.66 g/l washed yeast and 0.5 g/l talc, using flat sheet filters of polyvinylidene difluoride (PVDF) membranes. They found that when infrasonic frequency pulsing was applied the permeate, fluxes were four times higher for the talc suspension and three times higher for the yeast suspension.

CHAPTER 3

LITERATURE REVIEW OF THE APPLICATION OF ULTRASONIC TECHNIQUES TO MEMBRANE SYSTEMS

3.1 INTRODUCTION

Various methods have been used to measure or monitor fouling in industrial and laboratory membrane applications.[10] These methods provide information about the behaviour and the progression of membrane fouling. The nondestructive and noninvasive ultrasonic technique is a comparatively inexpensive measurement technique for the investigation of membrane fouling. Moreover, it can successfully monitor the growth of fouling layers.[10-12, 79, 80]

The ultrasonic technique has been studied and used to monitor the growth of fouling layer by many groups. Peterson et al.[10] found that UTDR could be utilized for the real-time measurement of the changes in membrane thickness under high pressure operating conditions, and also found that this technique did not interfere with the collection of standard performance data, for example, the permeate flux. Mairal et al.[11-78] described the first systematic attempt to adapt and use ultrasonics for the noninvasive measurement of membrane fouling during RO desalination of calcium sulfate solutions. They found that UTDR is sensitive to any changes that occur on the surface of the membrane due to the formation of a fouling layer.

Recently Li and coworkers used UTDR to monitor the membrane fouling.[81-85] Li and Sanderson[81] described the application of the UTDR technique to continuous visualization of particle deposition and its removal from a nylon membrane during cross-flow MF. Their results showed that UTDR could be used to monitor the growth of the fouling layer which provides useful information on the fouling process. Li et al.[83] described the application of the UTDR technique to the measurement of membrane fouling in a MF system with paper mill effluent from a wastewater treatment plant. The results showed a correspondence between the UTDR signal response from the membrane and the growth of the fouling layer on the surface of the membrane.

Li et al.[82] also used UTDR to measure organic fouling during ultrafiltration with polysulfone membranes. They again found that the ultrasonic signal response could be used to monitor fouling layer formation and growth on the membrane surface. Sanderson et al.[9, 86] used UTDR as a technique for visualization of membrane fouling and cleaning in a RO system. The UTDR technique could detect fouling layer initiation and its growth on the membrane surface.

Koen[87] used UTDR as a visualization technique to provide real-time characterization of the fouling layer during RO desalination in a system using flat-sheet membranes. The results showed an excellent correspondence between the flux decline behaviour and the UTDR response from the membrane. He also found that UTDR could be used to visualize membrane compaction and fouling. Sikdar et al.[88] studied the fouling during microfiltration of natural brown-coloured surface water by the UTDR technique, with a nylon membrane. Again it was found that the fouling process and fouling layer growth on the membrane surface could be monitored by applying the UTDR technique.

3.2 ULTRASONIC RANGES

Ultrasonic waves have a frequency range above the human hearing range. Sound ranges can be divided into three frequencies (Figure 3.1). Typically the human hearing range is from 16 Hz to 20 kHz. The sound waves below the human hearing range are called infrasound waves. The sound waves above the human hearing range are called ultrasound or ultrasonic waves.

The uses of ultrasound are divided into two areas: The first is high frequency ultrasound, which is used to measure the sound velocity, for medical scanning and chemical analysis (1 – 10 MHz), and the second is high energy ultrasound, which is used for cleaning, plastic welding and chemical reactivity (10 – 100 kHz).[12, 89, 90]

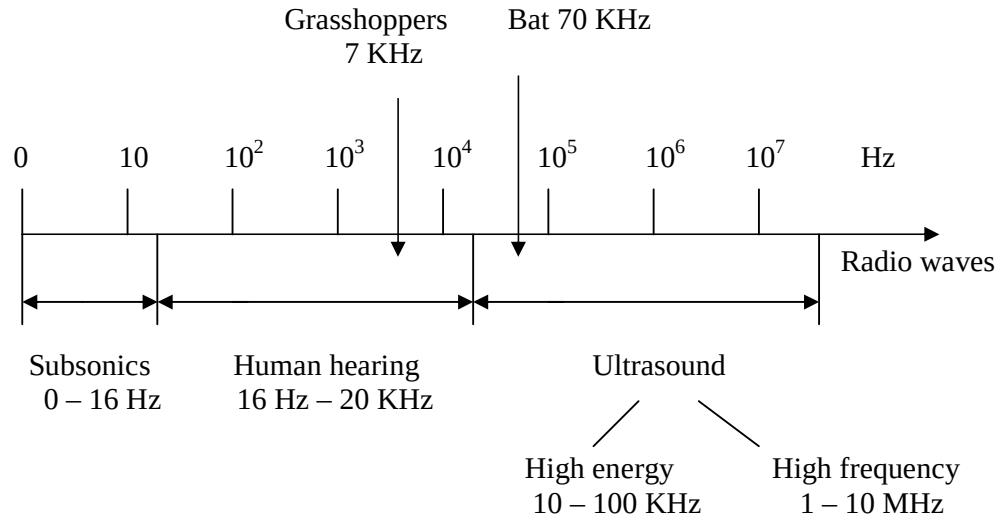


Figure 3.1 Diagram of sound frequencies.[90]

3.3 TYPES OF WAVES

An ultrasonic wave being transmitted in a material may be of different types. Each type results in a specific movement of the particles of the material in response to the wave. There are several types of waves used in ultrasonic testing, such as longitudinal waves, shear waves, surface waves and lamb waves.[91, 92] The different types of waves and their characteristics are given in (Table 3.1).[91]

Table 3.1 Types of waves[91]

Wave type	Direction of propagation	Characteristic velocity
Longitudinal	Same direction as or parallel to the direction of wave transmission	High velocities
Shear	Particle motion is perpendicular to the direction of wave transmission	Velocity almost half that of longitudinal waves

Lamb	Complex vibratory movements	Complex velocities in the direction of the vibratory movement
Surface	Elliptical particle motion, and can only be propagated on the material surface.	Velocity is about 90% of the shear waves velocity of the material

3.4 APPLICATION OF THE ULTRASONIC TECHNIQUE TO STUDY MEMBRANE FOULING

A cross-section view of a typical crossflow flat sheet membrane cell showing the principle of the ultrasonic technique measurement of fouling is illustrated in Figure 3.2. The cell consists of two polymethyl methacrylate (Perspex) plates. A membrane is placed between two Perspex plates. The transducer is mounted on top of the cell. This set-up was first described and used by Li et al.[83, 84] During the filtration process the feed solution flows over the top of the membrane and the permeate is withdraw from the bottom of the membrane. When fouling occurs on the membrane surface, the properties of the membrane change due to accumulation of foulants on the surface of the membrane. Because of this a fouling layer with thickness dS is present on the membrane surface, the reflected echoes A, B and C are produced from the different interfaces in the cell. Echo A is associated with the top plate/feed interface and echo B is associated with the initial feed solution/membrane interface.

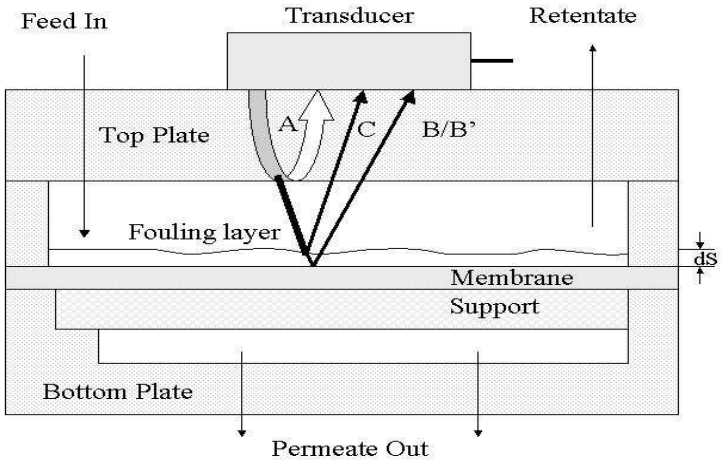


Figure 3.2 Schematic representation of the principle of ultrasonic technique measurement of fouling in a flat-sheet membrane cell.[81]

If the fouling layer is dense and thick enough to produce a reflected ultrasonic signal, a new echo signal will appear as a consequence of the new feed/fouling interface.[81, 83, 84] The corresponding time-domain response is illustrated in Figure 3.3.[84]

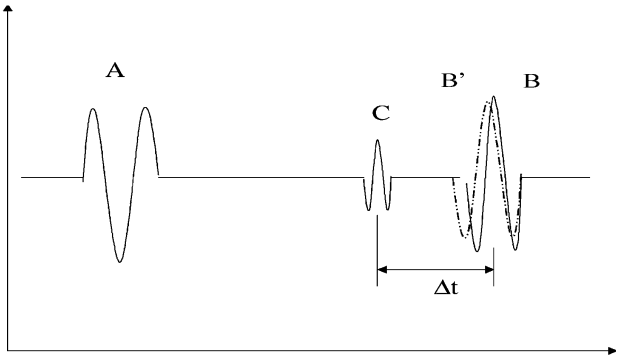


Figure 3.3 Corresponding time-domain response for set-up in Fig.12.[84]

The thickness of the fouling layer (dS) can be determined from the following equation:

$$dS = 0.5\ cdt \qquad (3.1)$$

where c is the ultrasonic velocity in the medium through which the wave travels and (dt) is the change in arrival time of the fouling peak.

CHAPTER 4

EXPERIMENTAL

4.1 INTRODUCTION

The backpulsing technique was applied to clean membrane fouling of MF and UF membranes. A washed yeast suspension was used as the feed solution during MF and UF processes with flat-sheet membranes; an alumina powder suspension was used as the feed solution during MF with flat-sheet membranes, and a dextrin suspension was used as the feed solution during UF with flat-sheet membranes.

MF and UF experiments were carried out using a flat-cell system. The system consisted of two parts connected to each other. The first part was the flat-cell filtration system and the second part was the ultrasonic measurement system. The ultrasonic technique was successfully applied to provide information about the growth of the fouling layer on the membrane surface and to monitor membrane cleaning.

After each experiment, the membrane was removed from the flat-cell and stored in a preservation solution (to prevent bacterial growth) prior to preparing the sample for examination of the surface properties, by SEM.

4.2 EXPERIMENTAL EQUIPMENT

4.2.1 FLAT-CELL FILTRATION SYSTEM

A schematic of the flat-cell membrane filtration system that was used for the MF and UF experiments is shown in Figure 4.1. A flat-cell membrane module is used to hold the membrane (Figure 4.2). The module is made of polymethyl methacrylate (Perspex) and made at the University of Stellenbosch. It has an effective membrane area of 0.0032 m^2. The module consists of two plates (each one 20 mm thick, 200 mm long and 94 mm wide), with a cavity in the top plate of 88 mm long, 30 mm wide and 13 mm deep. The membrane, covered by a spacer cloth, is clamped using an O ring between the two plates. There is a cavity in the topside of the top plate, its dimensions are: a side length of 100 mm, width 32 mm, and height

2.5 mm. The membrane was mounted on a brass support on the lower Perspex plate and below this was another cavity (88 mm long, 30 mm wide and 13 mm deep) to collect the permeate.

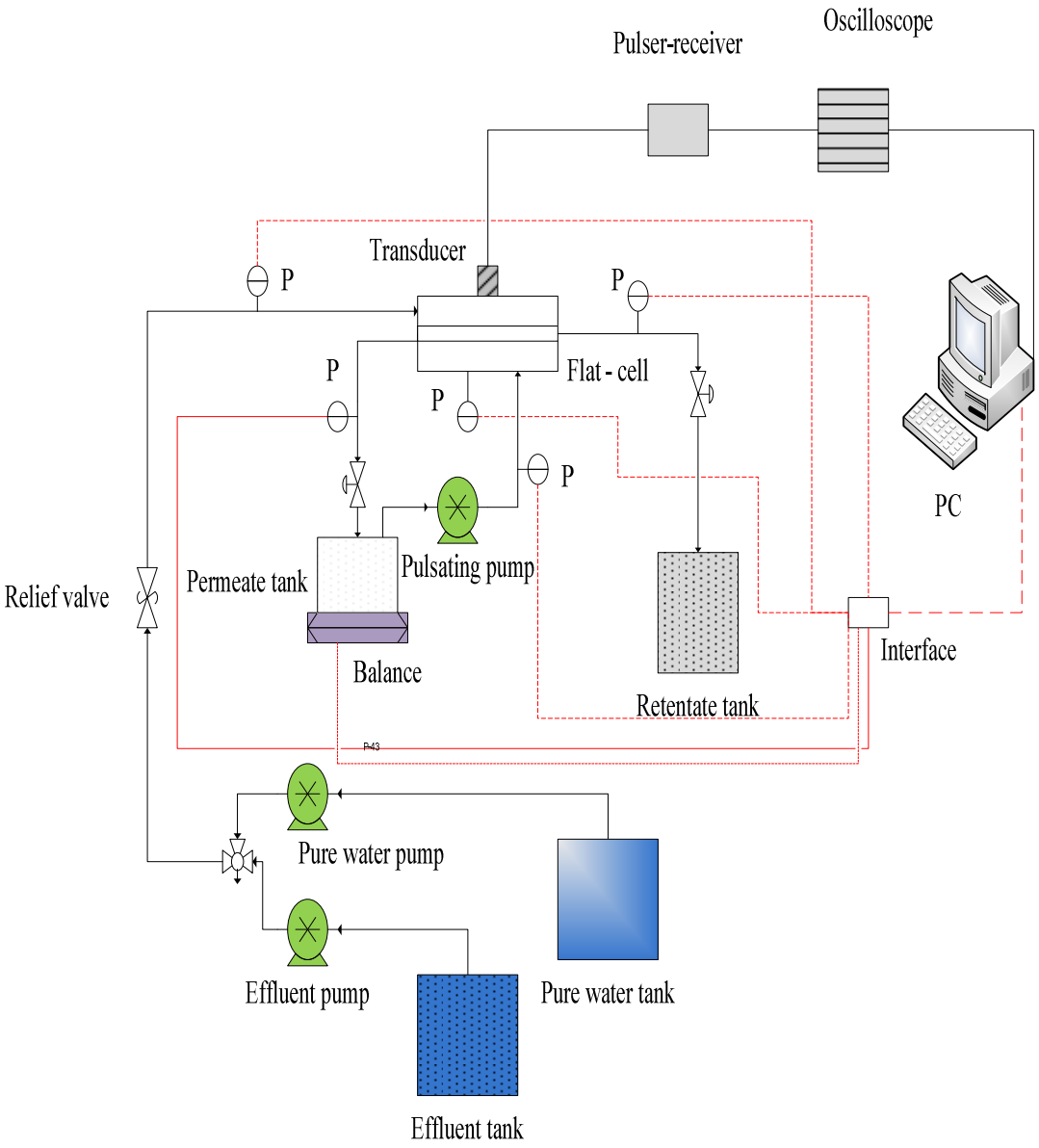


Figure 4.1 Schematic representation of experimental set-up of the flat-cell membrane filtration system.

The membrane module has two inlets and two outlets. The two inlets are for the feed inlet in the top plate and backpulsing inlet in the lower plate. The two outlets are for the retentate flow outlet in the top plate and for permeate flow in the lower plate.

Three pumps are connected to the system: two peristaltic pumps and a diaphragm pulsating pump. The two peristaltic pumps (Watson Marlow 323 and 313,) have a flow capacity of (0 – 0.86 l/min) at 0 – 400 rpm. The peristaltic pumps are connected to a single feed line by a three-way valve to feed either pure water or effluent into the membrane module.

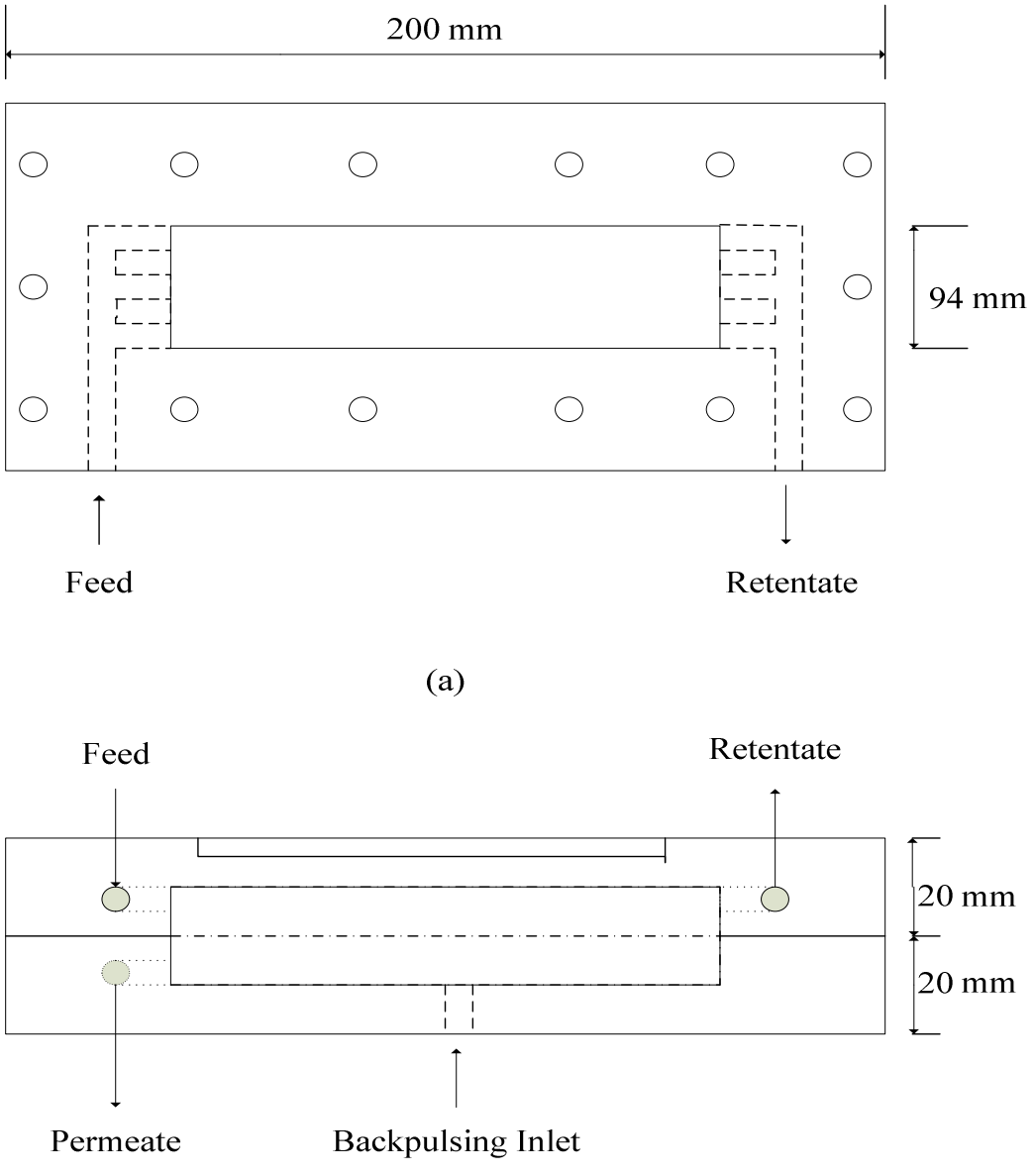


Figure 4.2 (a) Top view of the flat-cell membrane module, (b) side view of the flat-cell membrane module.[12]

One peristaltic pump is used to feed the flat-sheet membrane cell with pure water (RO water) to condition the membrane at constant pressure and the second peristaltic pump is used to feed the flat sheet membrane cell with effluent. A diaphragm pulsating pump (West Beach Instruments, Blouberg, RSA) has a constant pulse rate 400 of pulse/min (0 – 0.6 L/min) and was connected to the permeate side of the flat-cell membrane module. The frequency of the backpulsing was 5 to 6 Hz. It was the not objective of the research to optimize the frequency, but rather to establish the feasibility of pulsing in this frequency range. Figure 4.3 shows a typical pressure time trace of a pressure pulse. The permeate tank is used as a feed tank for the pulsating pump. Therefore either the pure water or the solution in the permeate tank can be used.

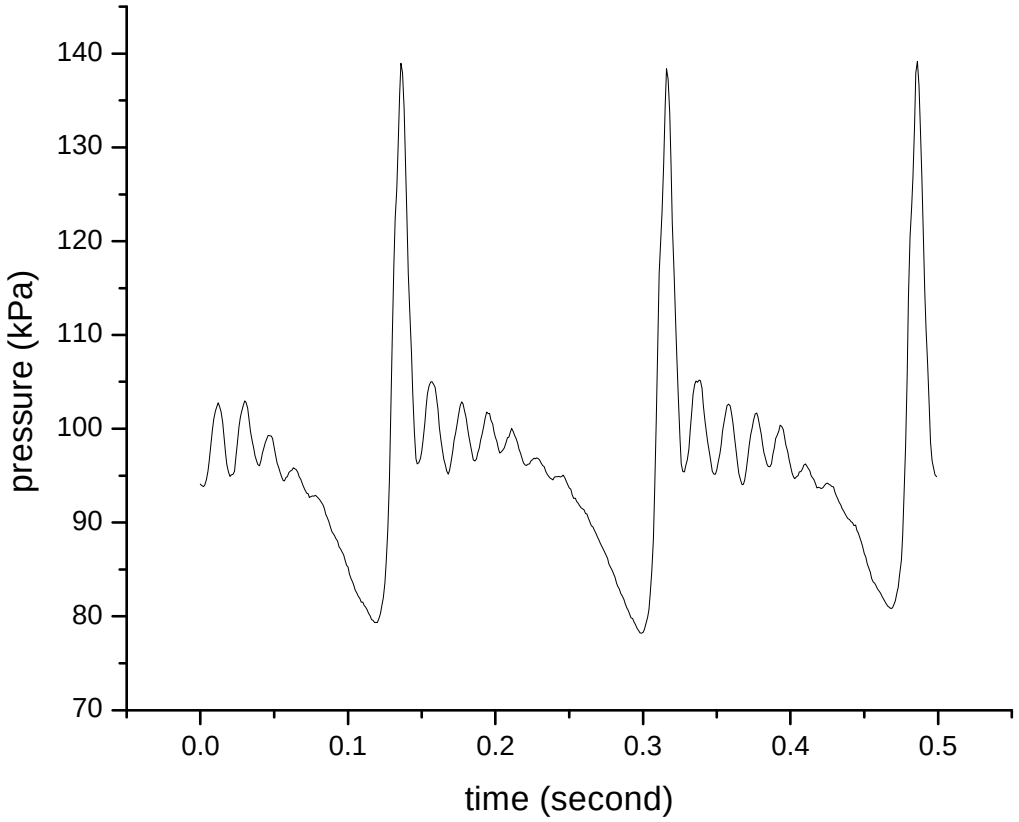


Figure 4.3 Pressure amplitude against time; in permeate space at the first couple of cycles of backpulsing.

A pressure relief valve is used in the feed line to maintain a constant feed pressure and also to provide protection from overpressure in the system. Pressure transmitters (WIKA Instruments, LR 110686-1, Milnerton, SA) with a 0 – 10 bar pressure range and 4 – 20 mA output are used to measure the feed, retentate and permeate pressures. The permeate flux was measured using an electronic balance and the mass change per unit time.

The pressure transmitters and the electric balance were all connected to a computer, which monitors the entire system using the graphic software called LabVIEW that allowed the measurements to be displayed on the computer screen. This program was used with labVIEW® 8.1 software (NI Solutions (Pty) ltd, Midrand) see Appendix 1.

4.2.2 ULTRASONIC MEASUREMENT SYSTEM

The ultrasonic measurement system (Figure 4.3) consisted of a transducer (7.5 MHz, Panametrics V120), a pulser-receiver (Panametrics 5058 PR), and a digital oscilloscope (HP model 54602B). The transducer was used to send and receive an ultrasonic pulse to and from the oscilloscope. The oscilloscope, that was connected to the pulser-receiver, captured the wave signal and displayed it as amplitude changes as a function of arrival time. A high viscosity ultrasound gel is used to couple the transducer to the surface. The oscilloscope was connected to the computer for data storage.

4.2.2.1 Pulser-receiver

A high voltage pulser-receiver (Panametrics 5058 PR) was used to generate the required voltage signals that enabled the transducer to send out an ultrasonic signal wave. The pulser-receiver was designed for ultrasonic testing and measurements applications that require a high material penetration capability. The Panametrics 5058 PR had a maximum excitation pulse

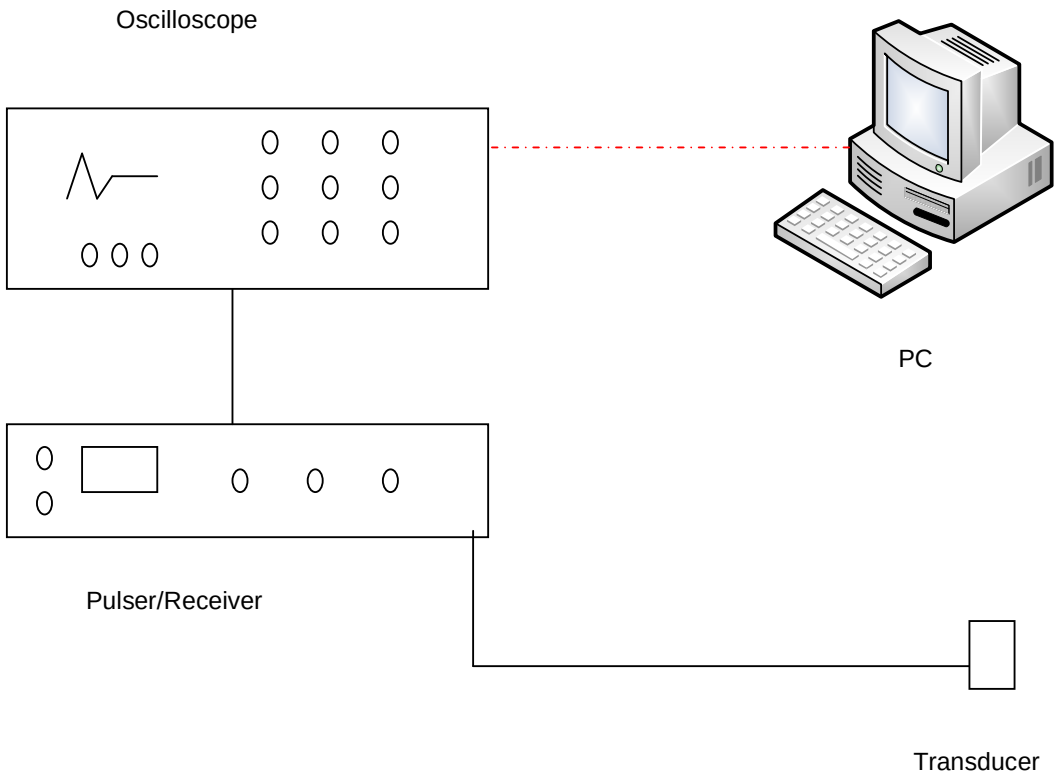


Figure 4.4 Schematic representation of experimental set-up of the ultrasonic measurement system

up to 900 volts. A 60 dB RF gain was provided by the receiver section, with an additional 30 dB available from an integral auxiliary preamplifier. The receiver gain was selectable at 40 or 60 dB. The receiver attenuation was adjustable from 0 dB to 80 dB in 1 dB steps. The front panel meter was used to display the maximum applied voltage to the transducer.

The settings of parameters of the pulser-receiver used in these experiments were as follows: pulse height 200 V, attenuator 25 dB, repetition rate 200 Hz, bandwidth 10 MHz gain 60 dB, and damping 50 ohms. The high pass filter and the low pass filter were not used, and the mode was normal.

Figure 4.5 Photograph of the Panametrics 5058 PR Pulser-receiver.

4.2.2.2 Oscilloscope

A digital Tektronix TDS 2024 oscilloscope was used to capture the wave signal produced and displayed it as amplitude changes as a function of arrival time. The settings of parameters of the oscilloscope were as follows.

- bandwidth 200 MHz
- sweep speed 5 s/div to 2 ns/div
- vertical sensitivity 1 mV/div
- up to 150 million samples / second

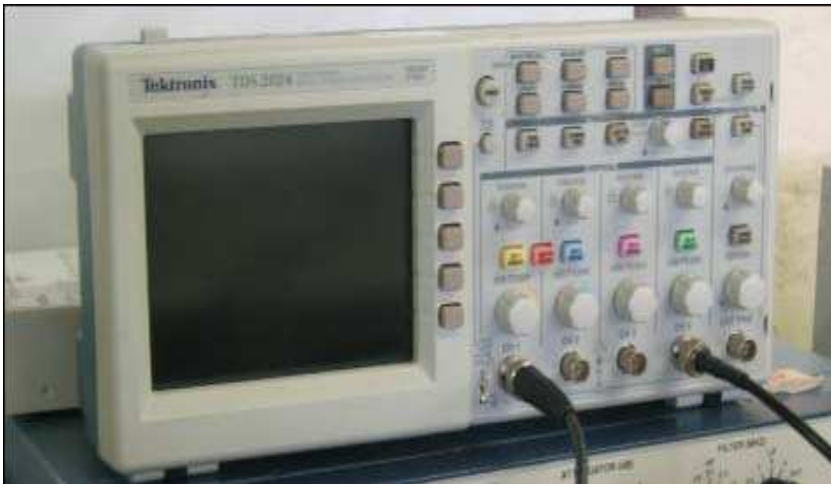


Figure 4.6 Photograph of the digital Tektronix TDS 2024 oscilloscope used in this study.

4.2.2.3 Ultrasonic transducer

A Panametrics videoscan transducer V120-RB with a central frequency of 7.5 MHz was used in this study. This provides a heavily damped broadband performance. Li et al.[81] had shown that it was the best choice for the system under investigation. The transducer was used to send an ultrasonic signal wave. It was mounted with a bracket into the cavity on the top of the flat-sheet membrane cell. A high-viscosity ultrasound gel was used to couple the transducer to the surface.

Figure 4.7 Photograph of the panametrics transducer V120-RB used in this study.

4.3 MEMBRANES AND FEED EFFLUENTS

4.3.1 MEMBRANES

4.3.1.1 MF MEMBRANES

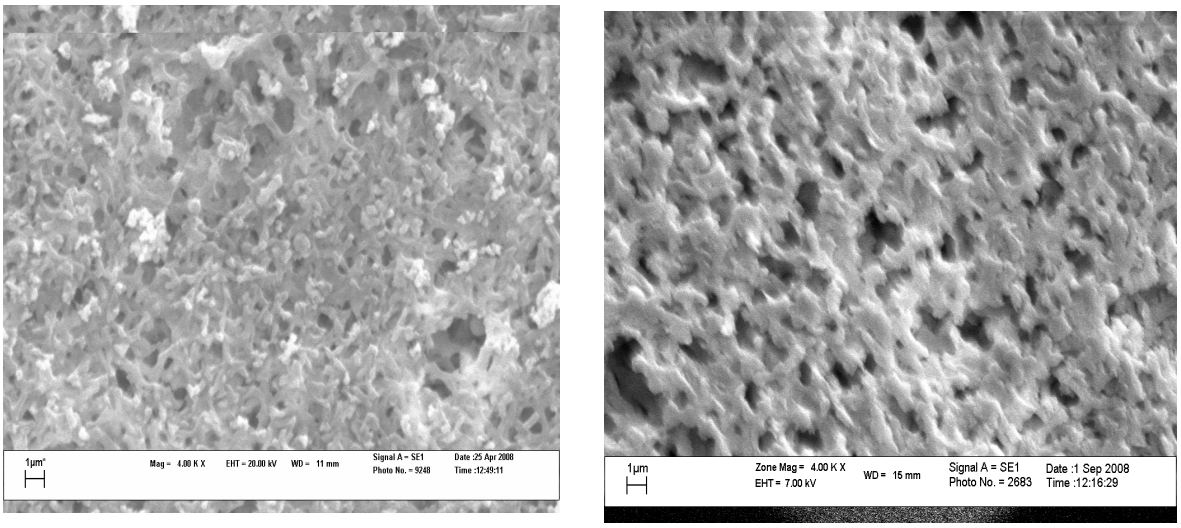
All MF experiments were carried out using one of two Biodyne A (amphoteric nylon 6,6) membranes (Pall Corporation, Pensacola, FL, USA). Biodyne membranes are composite layer membranes. The first membrane had a nominal pore size of 0.45 µm, the average thickness of the porous top layer was 60 µm, the nylon support layer was 50 µm and the porous bottom layer was 40 µm. The second membrane had a nominal pore size of 0.2 µm the average thickness of the porous top layer was 45 µm, the nylon support layer was 50 µm and the porous bottom layer was 55 µm. Each membrane was used once and then discarded. Membrane samples were cut from manufacturer-supplied rolls, several meters long and 30 cm wide.

SEM was used as analytical technique for the morphological characterization of the membranes surfaces. A series of SEM images were taken of new Biodyne membranes surfaces. Figure 4.8 shows a typical structure of the membranes surface, including the membranes pores. These SEM images of new membranes can be used as basic images to compare later with the fouled membranes.

4.3.1.2 UF MEMBRANES

All UF experiments were carried out using flat-sheet polysulfone (PS) membranes (GR40PP Alpha Laval, USA) with 100,000 molecular weight cut-off (MWCO) The support material is a polypropylene non-woven with thickness around 180 µm and the membrane layer is PS with thickness around 50-60 µm.

Membrane samples were cut from manufacturer-supplied rolls, several meters long and one meter wide. A series of SEM images were taken of the new PS membranes surfaces (see Figure 4.9).



(a) **(b)**

Figure 4.8 SEM micrographs of new Biodyne A (nylon) membranes: (a) 0.2 µm membrane, (b) 0.45 µm membrane. (Magnification 4,000X).

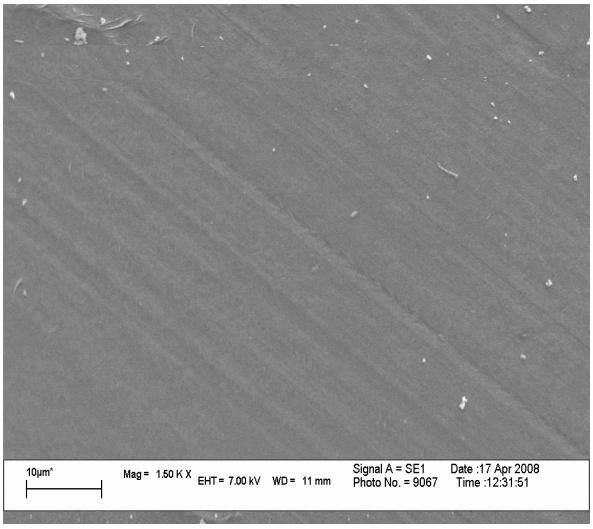


Figure 4.9 SEM micrograph of a new PS membrane. (Magnification 4,000X).

4.3.2 FEED EFFLUENTS

4.3.2.1 DEXTRIN SOLUTION

Dextrin solution was selected as the feed effluent which was used for UF experiments. Dextrin was purchased from Sigma Aldrich with molecular weight between 500 and 1000. The dextrin solution was made up of 1 L pure water (RO water) with 0.5 g dextrin (0.5g/L). The dextrin solution was selected because it is an organic molecular solution that represents a class of foulants often found in industry.

4.3.2.2 YEAST SUSPENSION

The effluent suspension was made up of pure water (RO water) with live commercial yeast cells. Before use, the yeast was washed by placing 1 g yeast in 60 ml of pure water, shaking well and then centrifuging the suspension in an Eppendorf Centrifuge 5702 at 2000 rpm for eight minutes. Then the clouded liquid portion was removed. This washing procedure was repeated four times. Then the dry mass of the washed yeast was used to make yeast suspensions. A yeast suspension concentration of 1 g/L was used for experiments with MF and UF membranes. The yeast suspension was selected because it forms a relatively adhesive cake layer.

4.3.2.3 ALUMINA SUSPENSION

The effluent suspension was made up of pure water (RO water) and alumina powder, the alumina was first washed by placing 1 g alumina in 60 ml pure water, then the mixture was shaken well and centrifuged in an Eppendorf Centrifuge 5702 at 1500 rpm for 5 minutes, then the liquid portion was removed. The remaining dry mass was used to make alumina suspensions. An alumina suspension concentration of 1 g/L was used for experiments with MF membranes. The alumina suspension was selected because it forms a relatively nonadhesive cake layer.

The dextrin solution, yeast suspension and alumina suspension were selected to test different types of foulants.

4.4 PROCEDURE

4.4.1 EXPERIMENTS USING A 90 KPA, 140 KPA, 180 KPA SEQUENCE

All the fouling experiments were made in cross flow mode. The operating feed pressure was ≈100 kPa with a feed flow rate of about 0.045 L/min.

During the beginning of this work preliminary experiments were performed to investigate which pulse amplitude from the diaphragm pulsating pump would give the best results. First the system (see Figure 4.10) was run with pure water (RO water) for about 15 minutes at 90 kPa (A) to obtain the pure water permeate flux. Then, the feed flow was changed from pure water to the effluent solution to initiate the fouling of the membrane. The membrane was fouled for 60 minutes at 100 kPa (B). Then, the effluent solution was replaced by pure water for 30 minutes at 100 kPa (C) to 90 minutes. The pulsating pump was then switched on for 35 minutes (D) with a peak pressure obtained from the oscilloscope trace of approximately 90 kPa (low), during this period the pulsating pump was switched off (for 1 minute) from time to time to enable the system to measure the true flux. The pulsating pump was then switched off for 15 minutes (E) to measure the new pure water permeate flux. After this the pulsating pump was switched on again for 35 minutes (F) with a peak pressure 140 kPa (medium). The pulsating pump was switched off again from time to time for measurements. The pulsating pump was then switched off again for 15 minutes (G) to measure the new permeate flux. The pulsating pump was switched on again for 35 minutes (H) with a peak pressure 180 kPa (high). Finally the pulsating pump was then switched off again for 15 minutes (I) to measure

the new permeate flux. The experimental procedure was done in this way to investigate which backpulse amplitude give the best results for permeate flux values.

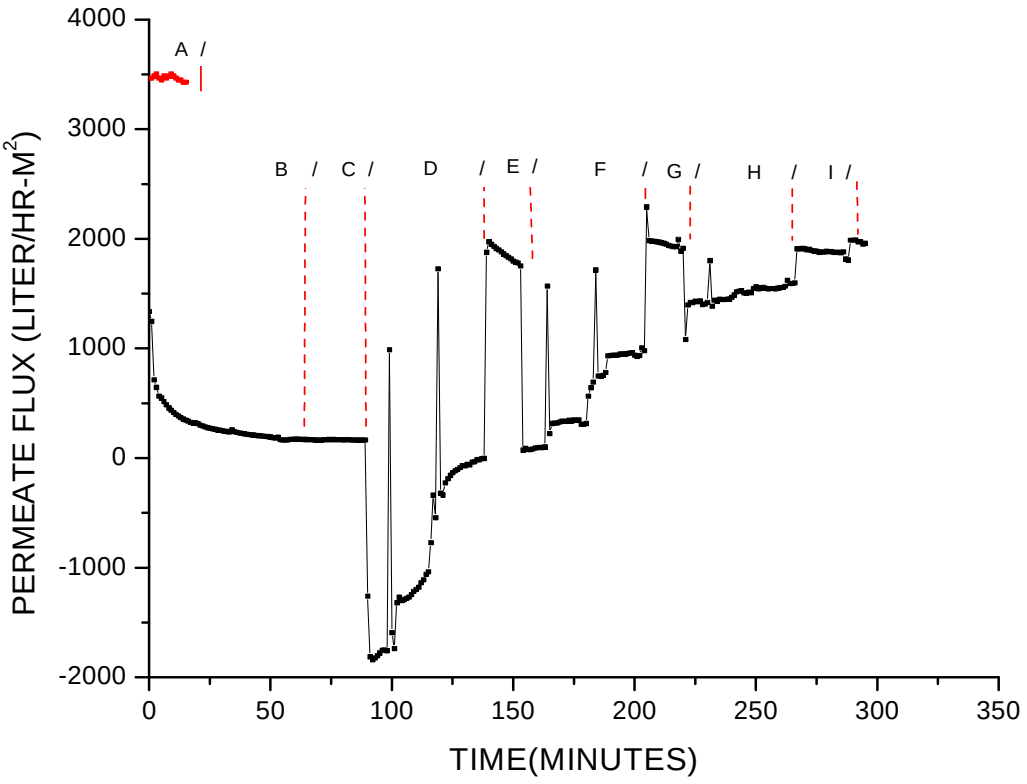


Figure 4.10 Diagram illustrating experimental procedure.

4.4.2 **DEFOULING EXPERIMENTS USING A THREE 140 KPA PULSE SEQUENCE**

The fouling experiments were operated in cross flow mode. The operating feed pressure was ≈100 kPa with a feed flow rate of about 0.045 L/min. the time sequence for these experiments and the experiments in 4.4.1 were the same.

Initially, in each experiment (see Figure 4.10) pure water (RO water) was used as the feed water, at the same feed pressure and flow rate for about 15 minutes (A) to obtain the pure water permeate flux. Then, the feed flow was changed from pure water to the effluent solution for the fouling of the membrane. The membrane was fouled for 60 minutes (B) where the membrane reached a near steady-state flux. The effluent solution was replaced by pure water for 30 minutes(C), the pulsating pump was switched on for 35 minutes (D) with a peak pressure, obtained from the oscilloscope trace, of approximately 140 kPa, using pure water in

the permeate tank as a source to the pulsating pump. During this period the pulsating pump was switched off from time to time to enable the system to measure the true flux, then at the end of the cycle the pulsating pump was switched off for 15 minutes (E) to measure the new pure water permeate flux. Then the pulsating pump was switched on again for 35 minutes (F) with the same peak pressure 140 kPa, using pure water or a SES soap solution* or a F9 soap solution** as the source for the pulsating pump, again the pulsating pump was then switched off again from time to time. The pulsating pump was switched off again at the end of the cycle for 15 minutes (G) to measure the new permeate flux. The pulsating pump was switched on again for 35 minutes (H) with the same peak pressure 140 kPa using pure water as a source to the pulsating pump. The pulsating pump was switched off again for 15 minutes (I) to measure the new permeate flux.

* SES soap solution was selected as one of the cleaning solution; the solution was made of 1 L pure water with 1 g ethylene diamine tetraacetic acid (EDTA), 1 g sodium lauryl sulfate (SLS) and 1 g calcium hypochloride.

** F9 soap solution was selected as one of the cleaning solution; the solution was made of 1 L pure water with 1 g of nonylphenol ethoxylate.

4.4.3 ULTRASONIC MEASUREMENTS

UTDR was used to monitor the growth of fouling layers on the membrane surfaces and provided information about the efficiency of backpulsing cleaning (described in Section 3.4). The transducer was used to send and receive an ultrasonic pulse to and from the oscilloscope. The ultrasonic measurement system captured the changes in ultrasonic signal responses (every minute). These data were stored on the computer. Ultrasonic measurements were taken on the new membrane at 0 second ($t = 0$) in each experiment and used as a baseline measurement. The ultrasonic data obtained from the experiments were analysed to ultrasonic differential and hence determination of differential signals from the recorded baseline measurement.

4.4.4 MEMBRANE ANALYSIS

After each defouling experiment the cleaned membrane was removed from the flat-cell filtration and stored in a glass jar containing preservation solution, the solution was made of 1 g sodium metabisulphite with 1 L pure water (1 g/L). In addition, some special experiments were performed for 15 seconds and 60 minutes, and then these membranes were removed from the flat-cell filtration and stored again in the preservation solution. Small sections were cut from the stored membrane and prepared for SEM and then analysed using a Leo® 1430VP Scanning Electron Microscope. The samples were coated with gold just prior to imaging or analysis to prevent electron charging effects on the sample. The beam conditions during analysis were 20 KV and approximately 1.5 nA, with a working distance of 13 mm.

CHAPTER 5
RESULTS AND DISCUSSION

5.1 EXPERIMENTS USING A 90 KPA, 140 KPA, 180 KPA SEQUENCE

All experiments were carried out using three different effluent solutions/suspensions, an alumina suspension, a yeast suspension and a dextrin solution using Biodyne A (amphoteric nylon 6, 6) 0.2 μm membrane, Biodyne A (amphoteric nylon 6, 6) 0.45 μm membrane and 100,000 MWCO PS membrane at the same operating conditions, at feed pressure 100 ± 3 kPa with a flow rate of 0.045 ± .003L/min, while the temperature was kept constant at 25 ± 1◦C.

Results obtained from the sequential backpulsing experiments are shown in Figures 5.1 – 5.3. Figure 5.1 shows a result of the flux against time when an alumina suspension is used with a Biodyne A (amphoteric nylon 6, 6) 0.2 μm membrane during a MF process. The initial flux rapidly decreased over the first 20 minutes of operation, followed by a gradual decrease until 60 minutes of operation. The pure water flux increases noticeably after the low pressure pulse at 135 minute, and the medium and high pressure pulse have cleaned the membrane to 4658 and 4668 $L.h^{-1}.m^{-2}$ at 185 and 235 minute, respectively, which are 99.7% and 99.9% of the initial value (pure water value) of 4699 $L.h^{-1}.m^{-2}$, respectively.

Figure 5.2 shows that when a washed yeast suspension was used, with a Biodyne A (amphoteric nylon 6, 6) 0.45 μm MF membrane, there is a rapid decline in permeate flux after 15 seconds. This rapid decline in permeate flux is due to the deposition of yeast particles on the membrane surface and the membrane pores becoming blocked. The low pressure pulse has virtually no effect on cleaning of the membrane, but the medium and high pressures pulses cleaned the membrane to fluxes of 2495 and 2650 $L.h^{-1}.m^{-2}$ respectively, which are 67% and 71% of the initial flux value (3720 $L.h^{-1}.m^{-2}$).

Figure5.3 shows that when a washed yeast suspension was used with the 100,000 MWCO PS UF membrane, after the low, medium and high pressure pulses, the flux values are 70, 90 and 100 $L.h^{-1}.m^{-2}$ respectively, which are 62%, 80% and 88% of the initial value (113 $L.h^{-1}$.m-2). Only the 0.45 μm Biodyne nylon membrane shows little difference at 140 and 180 kPa feed pressure. The 0.2 μm Biodyne nylon and PS membranes need the 180 kPa feed pressure for adequate flux restoration.

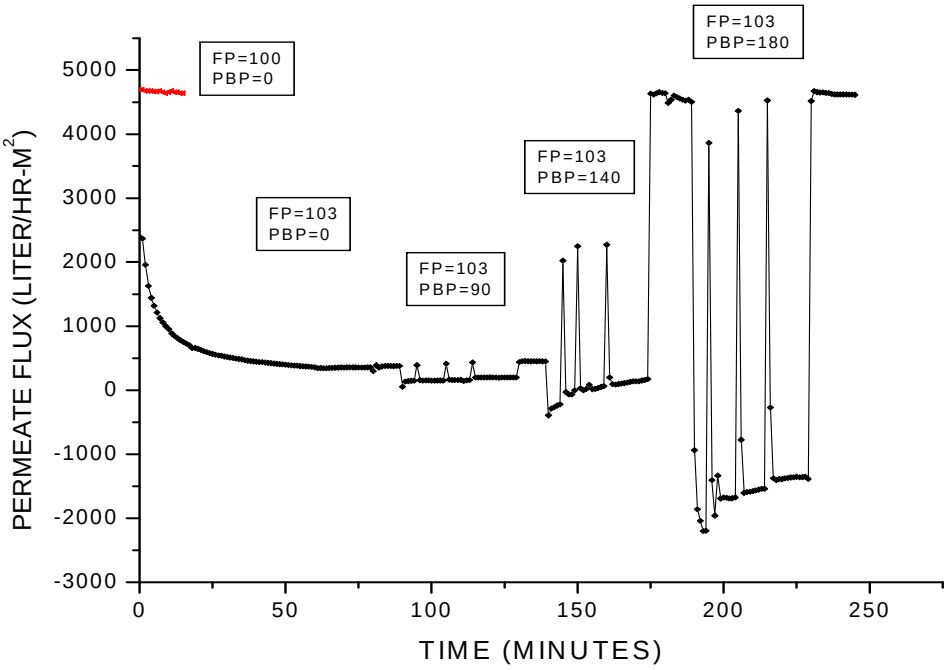


Figure 5.1 Flux against time for the Biodyne A (amphoteric nylon 6, 6) 0.2 μm membrane /alumina system. (FP: feed pressure, PBP: peak of backpulse pressure).

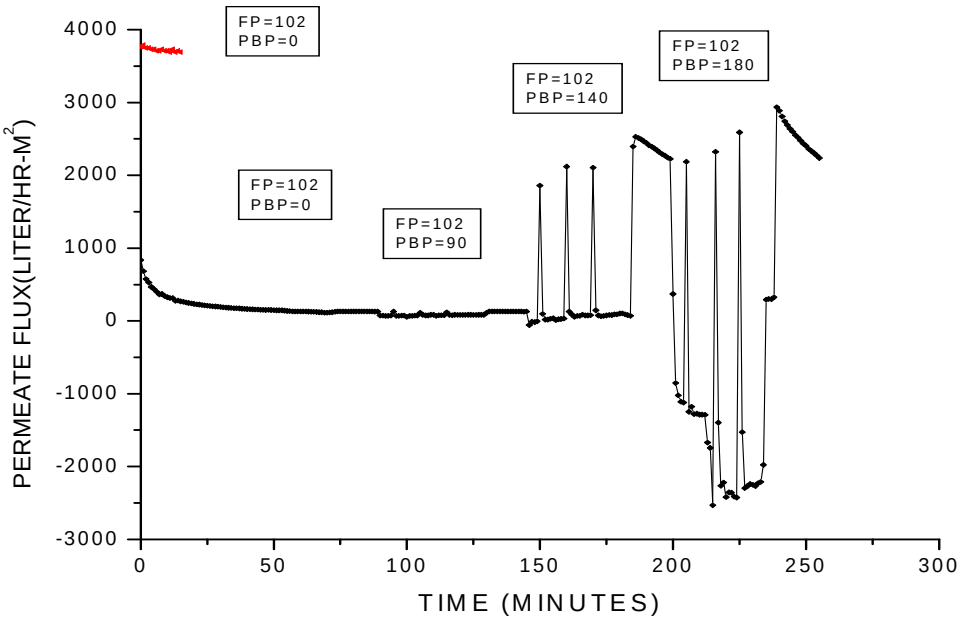


Figure 5.2 Flux against time for Biodyne A (amphoteric nylon 6, 6) 0.45 μm membrane /yeast system. (FP: feed pressure, PBP: peak of backpulse pressure).

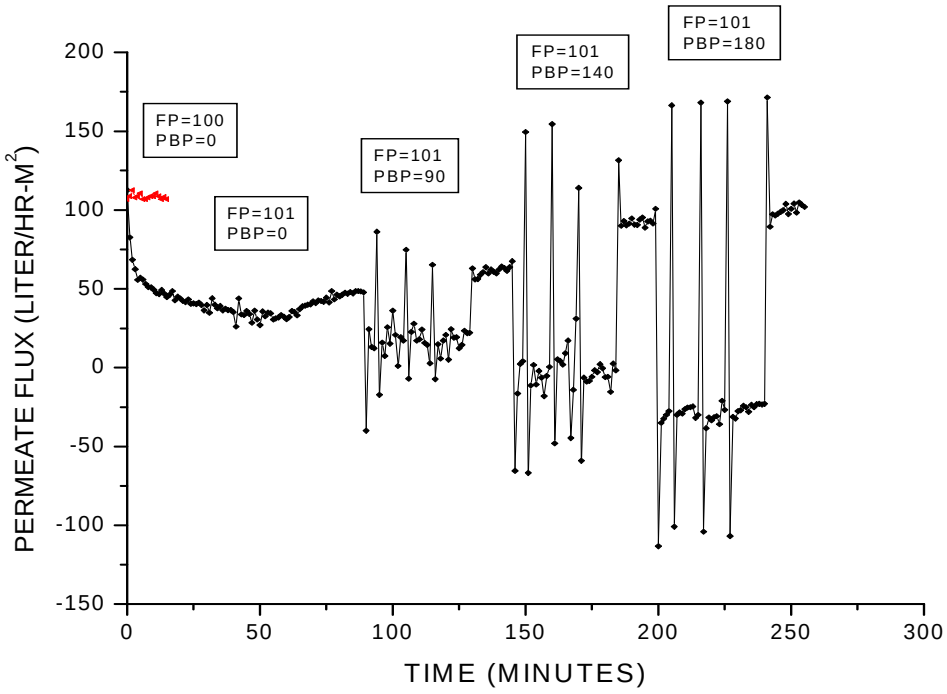


Figure 5.3 Flux against time for the 100,000 MWCO PS membrane /yeast system. (FP: feed pressure, PBP: peak of backpulse pressure).

5.2 FOULING WITH AN ALUMINA SUSPENSION IN A MF SYSTEM

5.2.1 DEFOULING EXPERIMENTS USING A THREE 140 KPA PULSE SEQUENCE

All experiments in this section were carried out using Biodyne A (amphoteric nylon 6, 6) 0.2 μm membrane. The membrane was fouled using an alumina suspension during a MF process. The test conditions for experiments are listed in Table 5.1.

Table 5.1 Test conditions used for the alumina defouling experiment

Parameter	Value
Feed flowrate	0.045 ± .003L/min
Feed pressure	100 ± 3 kPa
Temperature	25 ± 1◦C
Feed concentration	1 g/L
pH	8.5

Figure 5.4 shows a plot of flux as a function of operating time for the alumina/ 0.2 μm nylon membrane defouling experiment using pure water as feed solution only for the backpulsing pump (the reproducibility is shown by error bars based on two experiments). The flux rapidly decreases in the first 20 minutes of operation followed by a gradual decrease until 60 minutes of operation, and then the flux becomes steady, from 60 to 90 minutes during the pure water wash. During the first cleaning pulse negative flux values are visible. After this a new pure water flux value was measured for 15 minutes, which showed that the first cleaning pulse cleaned the membrane up to a flux value of 2560 $L.h^{-1}.m^{-2}$ at 135 minute, which is 87% of the initial value (2960 $L.h^{-1}.m^{-2}$). The second cleaning pulse cleaned the membrane up to 2650 $L.h^{-1}.m^{-2}$ at 185 minute, which is 88% of the initial pure water flux value (2960 $L.h^{-1}.m^{-2}$). The third cleaning pulse cleaned the membrane up to 2890 $L.h^{-1}.m^{-2}$ at 235 minute, which is 95% of the initial value (2960 $L.h^{-1}.m^{-2}$).

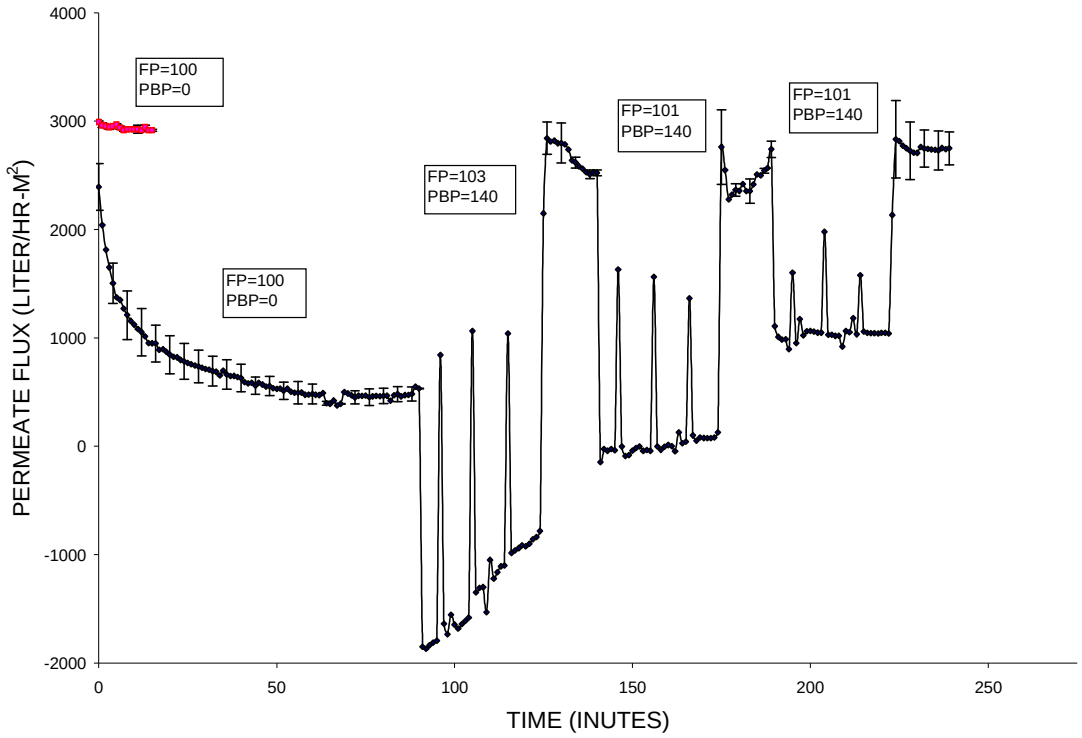


Figure 5.4 Flux against time for the Biodyne A (amphoteric nylon 6, 6) 0.2 µm membrane /alumina system (all backpulsing with pure water). (FP: feed pressure, PBP: peak of backpulse pressure).

Figure 5.5 shows the plot of flux as a function of operating time for the alumina/ 0.2 µm nylon membrane defouling experiment, using soap solution (SES solution) as feed solution for the backpulsing pump during the second cleaning pulse. The first cleaning pulse cleaned the membrane flux to 3140 $L.h^{-1}.m^{-2}$ at 135 minute, which is 98% of the initial value (3170 $L.h^{-1}.m^{-2}$). The second cleaning pulse cleaned the membrane to 3145 $L.h^{-1}.m^{-2}$ at 185 minute, which is 98% of the initial value (3170 $L.h^{-1}.m^{-2}$). The third cleaning pulse cleaned the membrane to 3155 $L.h^{-1}.m^{-2}$ at 235 minute, which is 99% of the initial value (3170 $L.h^{-1}.m^{-2}$).

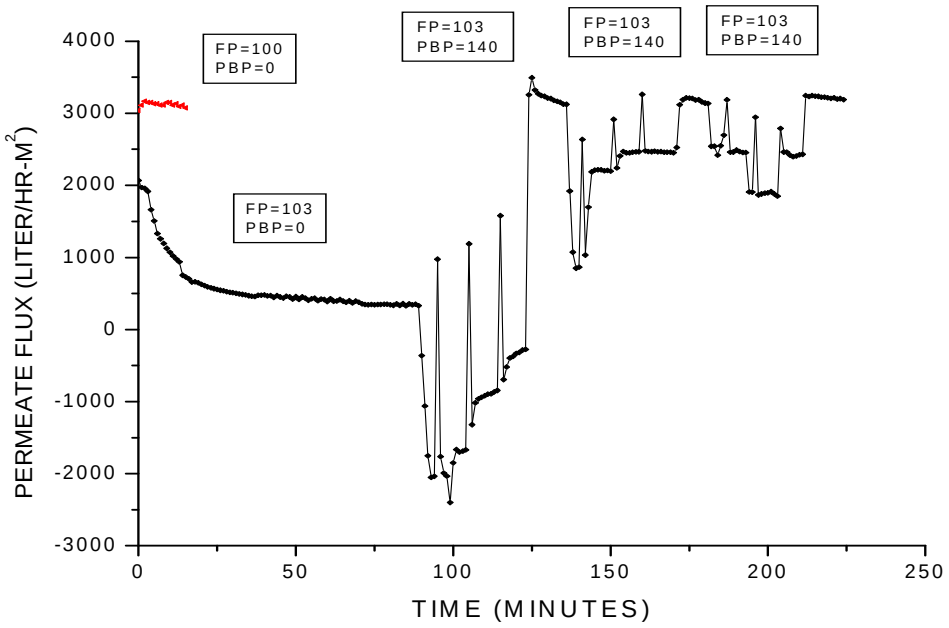


Figure 5.5 Flux against time for the Biodyne A (amphoteric nylon 6, 6) 0.2 μm membrane /alumina system (second backpulsing with soap solution). (FP: feed pressure, PBP: peak of backpulse pressure).

5.2.2 ULTRASONIC MEASUREMENTS

Figure 5.6 shows the complete ultrasonic signal obtained from the membrane inside the flat-cell membrane module during pure water operation with a new clean nylon membrane. It provides information about the basic signal of ultrasonic measurements. The first echo (A) is the reflection of the signal off the Perspex top plate of the cell and the water interface, the second echo (B) is the reflection of the signal off the water and the nylon membrane interface, and the third echo (C) is reflected from the interface of the nylon membrane and the porous metal support. The ultrasonic measurement is focused on changes in the echo B, which was used to calculate the thickness of the fouling layer in the fouling experiments.

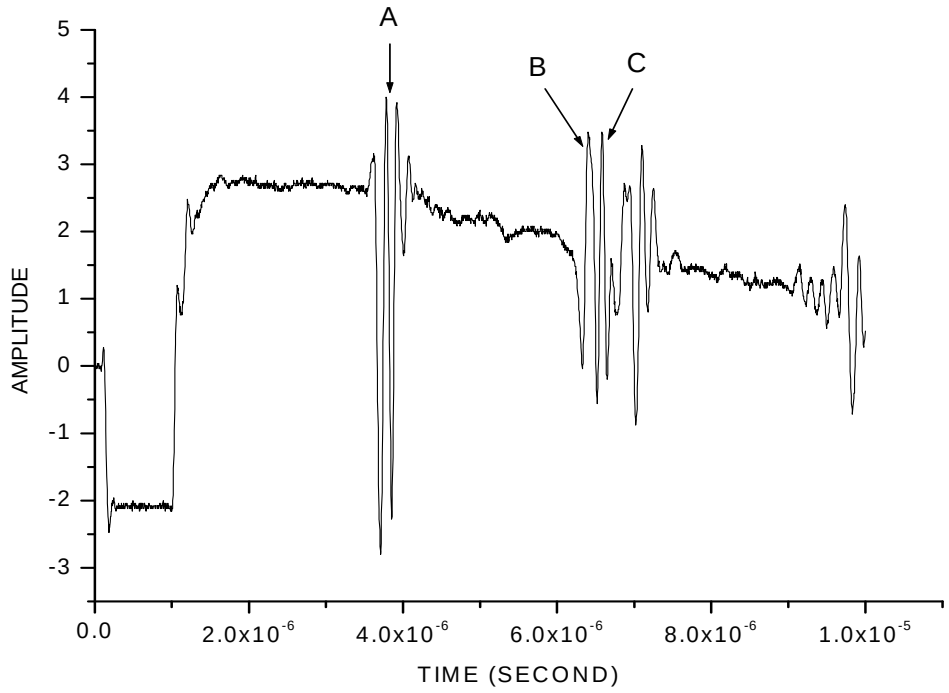


Figure 5.6 Ultrasonic spectrum of the flat-cell during pure water filtration (at 0 minute) using a new 0.2 µm nylon membrane.

Figure 5.7 is a cross-sectional view of the cell with received reflections from the various interfaces.

If the speed of sound in Perspex, water and a nylon membrane is known, their thicknesses can be measured using Equation 3.1 with

$C_{perspex}$ = 2730 m/s

C_{water} = 1438 m/s

$C_{polyimade\ (nylon)}$ = 2200 m/s.[89]

The arrival time of the response signals as measured by UTDR (Figure 5.6) was:

t_A = 3.63 µs

t_B = 6.42 µs

t_C = 6.56 µs

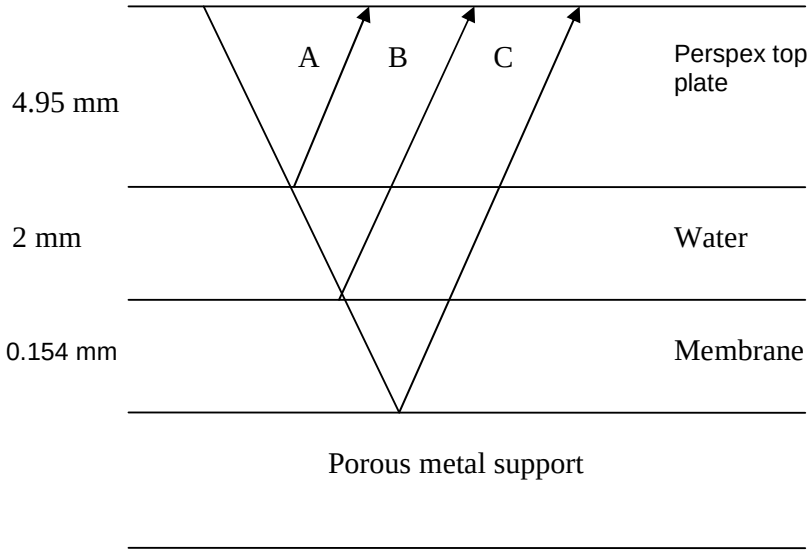


Figure 5.7 Cross-sectional view of the cell with received reflections from the various interfaces.

$$dS_{Perspex} = 0.5 \times C_{Perspex} \times t_A$$
$$= 0.5 \times 2730 \times 3.63$$
$$= 4.95 \times 10^{-3} \text{ m}$$
$$= 4.95 \text{ mm}$$
$$dS_{water} = 0.5 \times C_{water} \times (t_B - t_A)$$
$$= 0.5 \times 1438 \times (6.42 - 3.63)$$
$$= 2 \times 10^{-3}$$
$$= 2 \text{ mm}$$
$$dS_{nylon} = 0.5 \times C_{polyimade\ (nylon)} \times (t_C - t_B)$$
$$= 0.5 \times 2200 \times (6.56 - 6.42)$$
$$= 0.154 \times 10^{-3}$$
$$= 0.154 \text{ mm}$$

The calculated thickness of the Perspex plate is 4.95 mm, which is very close to the measured value of 5.0 mm by vernier caliper. The calculated thickness of the nylon membrane is 0.154, which is very close to the measured value of 0.15 mm. Overall, the model showed good correlation between the measured cell dimensions and the echoes received.

Figure 5.8 shows the changing amplitude of the reflected pulse recorded as a function of arrival times for the data given in Figure 5.4. The 0 minute signal shows the peak near 5.3 E^{-6} seconds generated from the pure water/new membrane interface and internal reflections from the membrane structure, and was taken just before switching to the alumina suspension feed. This takes about 20 seconds for the feed change over the membrane. Each signal had a number of defined peaks, which generated from different interfaces of the membrane layers and the support layers. This 0 minute signal is used as a reference signal for later use during fouling and cleaning process. The density of the water-saturated upper layer of the nylon membrane would be very similar to density of water. So, the first peak on 0 minute signal is likely to be resulted from the central nylon support of the new membrane, before the fouling began because of the very small change in density from water to membrane. After 10 minutes of fouling there is a water/foulant peak which becomes clearly visible in front of the membrane peak because of the formation of the alumina layer. This first peak is shifted towards earlier arrival times, up until 60 minutes, at which time the membrane is almost completely fouled. This is because of the gradual increase in the density and the thickness of the cake layer. Results of measurements (see section 5.2.3) showed that the thickness of the fouling layer was 375 µm, after 60 and 85 minutes, and that there was no effect on the thickness of the fouling layer after washing with pure water. The signal reflections at 135, 185 and 235 minutes, which are after first, second and third cleaning pulses respectively, show that the first cleaning pulse removes all or most of the fouling layer. It is also evident that the membrane properties (i.e. density) had changed due to some leftover particles on the surface (see Figure 5.9) of the membrane. This is supported by the remaining fouling layer thickness (Figure 5. 10).

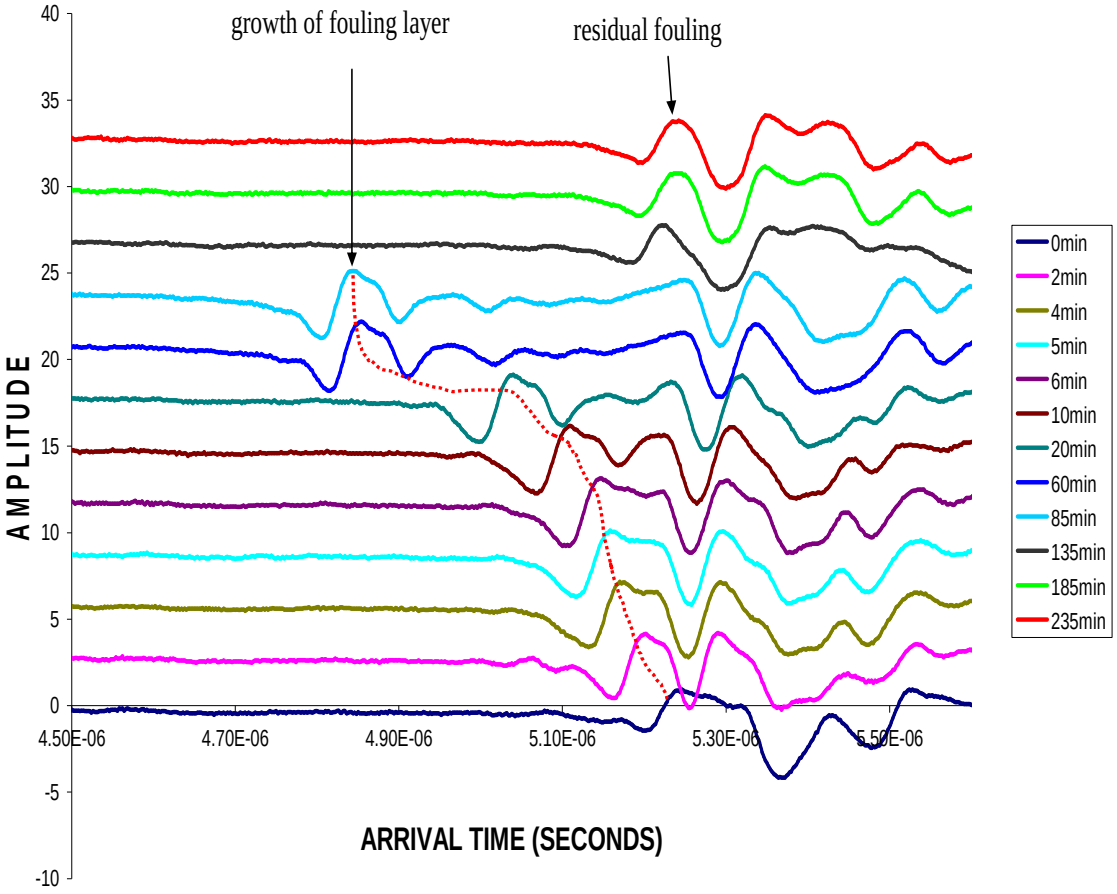


Figure 5.8 Amplitude of the reflection received at the detector as a function time, for the Biodyne A (amphoteric nylon 6, 6) 0.2 μm membrane /alumina system. The time interval shown encompasses all the reflections received for the water/film, film/membrane and membrane/metal support interfaces.

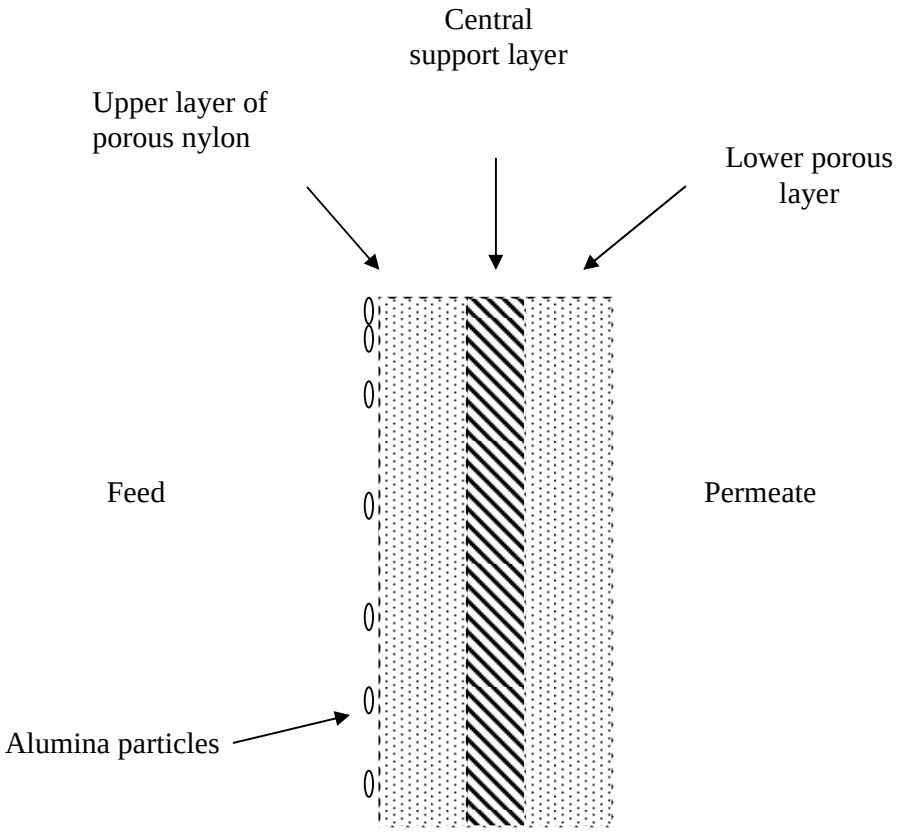


Figure 5.9 Proposed cross-section of the 0.2 µm nylon membrane cleaned by backpulsing (at 235 minutes).

5.2.3 DETERMINATION OF THE FOULING LAYER THICKNESS AS A FUNCTION OF TIME

The thickness of the fouling layer was calculated at each time by measuring the difference in arrival times between the reflection from the growing water/foulant interface and that from the water/new membrane interface, using the time-distance relationship in equation (3.1). The velocity of sound in the medium was 1530 m/s, established by Li et al.[82]

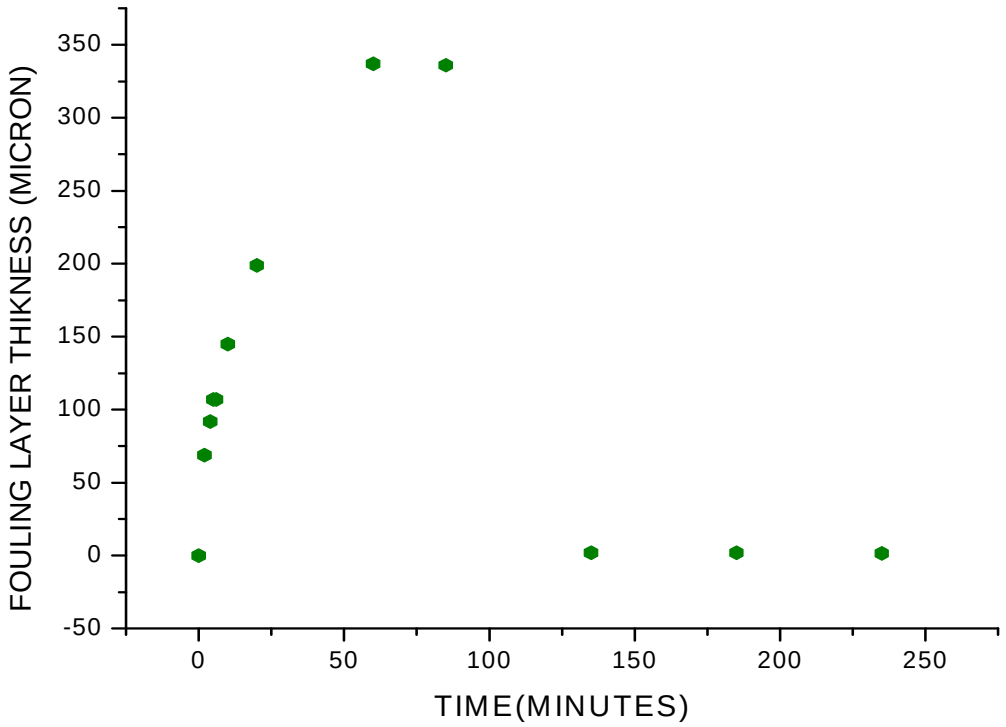
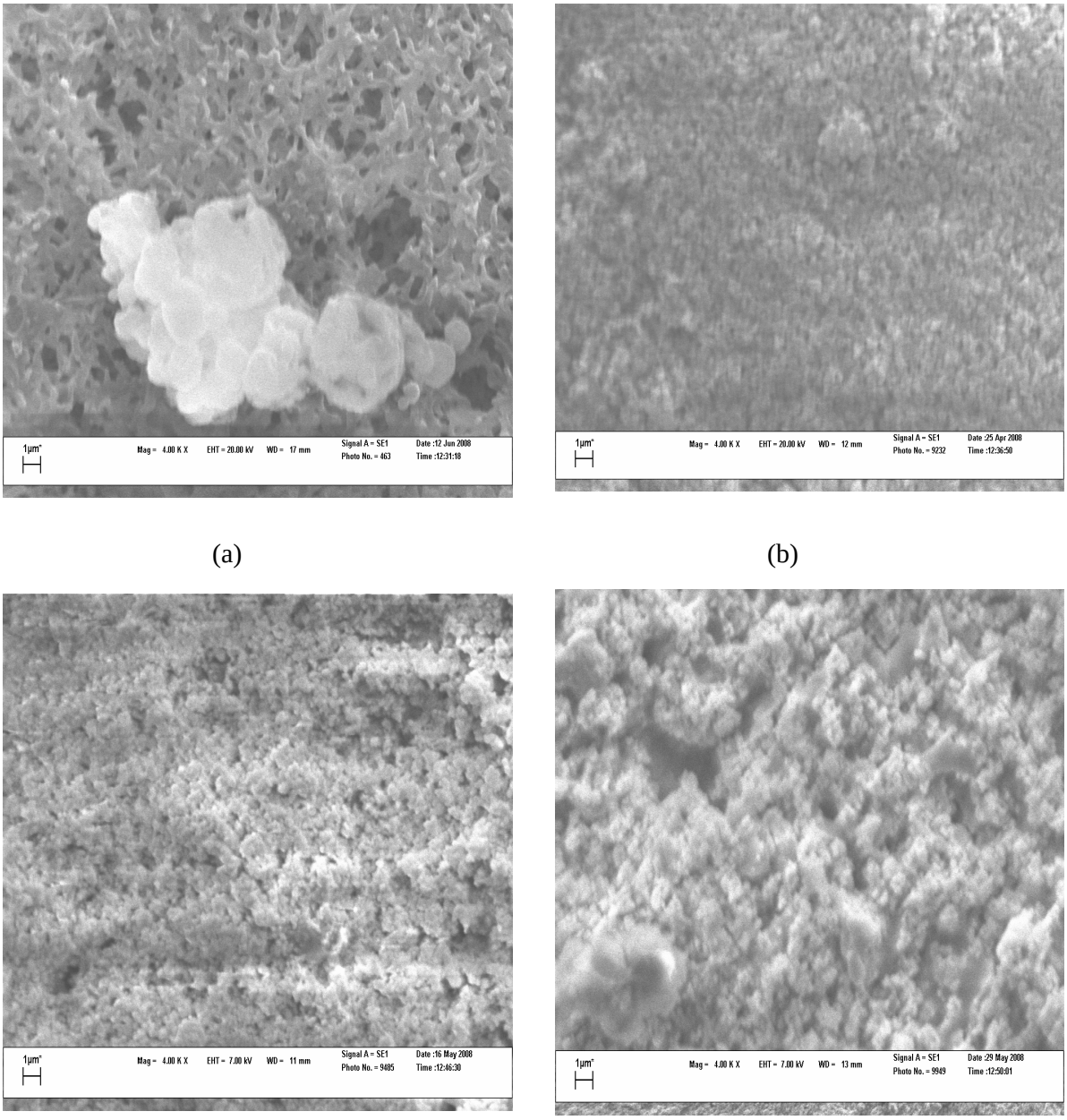


Figure 5.10 The fouling layer thickness of the Biodyne A (amphoteric nylon 6, 6) 0.2 μm membrane /alumina system as a function of time.

5.2.4 SEM ANALYSIS

Figure 5.11 shows SEM images (Magnification 4,000X) of the fouled and cleaned nylon 0.2 μm membranes for forward filtration experiments carried out with the alumina suspension. Figure 4.8 (a) has shown previously the new membrane surface structure, including the pores of the membrane. Figure 5.11 (a) shows an image of a fouled membrane taken after 15 seconds where the flux was 2620 $L.h^{-1}.m^{-2}$. This image shows no visible pore blocking, but some alumina particles starting to form an alumina cluster. Figure 5.11 (b) shows an image of a fouled membrane after 60 minutes of forward cross-flow filtration. The membrane is now completely covered by an alumina cake layer. Figure 5.11 (c) shows an image of a cleaned membrane after the third cleaning backpulse. Almost complete membrane cleaning is obtained in these experiments by the backpulsing cleaning method; for Figure 5.11 (c) the flux is slightly lower than the pure water flux. Figure 5.11 (d) shows an image of cleaned membrane taken after the third cleaning backpulse. Also almost complete membrane cleaning is obtained in these experiments using the backpulsing cleaning method. High magnification SEM is representative of the surface as indicated by lower magnification images (see Appendix 2).



(a) (b)

(c) (d)

Figure 5.11 SEM images (magnification 4000X) of the Biodyne A (amphoteric nylon 6, 6) 0.2 µm membrane /alumina system. (a) Membrane surface after being fouled for 15 seconds, (b) a fouled (60 minute) surface, (c) surface cleaned by three successive pure water backpulses, (d) surface cleaned by pure water, soap solution and pure water backpulses.

5.3 FOULING WITH A YEAST SUSPENSION IN A MF SYSTEM

5.3.1 DEFOULING EXPERIMENTS USING A THREE 140 KPA PULSE SEQUENCE

All experiments were carried out using a Biodyne A (amphoteric nylon 6, 6) 0.45 μm membrane. The membrane was fouled using a washed yeast suspension during MF. The test conditions used for the experiments are listed in Table 5.2.

Table 5.2 Summary of test conditions for yeast defouling experiment.

Parameter	Value
Feed flowrate	0.045 ± .003L/min
Feed pressure	100 ± 3 kPa
Temperature	25 ± 1◦C
Feed concentration	1 g/L
pH	8

Figures 5.12 and 5.13 show the plots of the forward filtration flux as a function of time for washed yeast defouling experiments with three cleaning cycles: backpulsing with pure water during the first, second and third cleaning pulses (Figure 5.12), and with pure water during the first, SES solution during the second and pure water during the third cleaning pulses (Figure 5.13). In all cases the flux decreases rapidly at first and more slowly at longer times until 60 minutes of operation, before becoming 175 $L.h^{-1}.m^{-2}$ at 60 minutes, which is 7% of the initial value (3465 $L.h^{-1}.m^{-2}$). After the pure water wash the flux values are somewhat lower. After three pure water cleaning pulses (Figure 5.12), the following pure water flux values were recorded after first cleaning pulse: 1990 $L.h^{-1}.m^{-2}$ at 135 minutes, this is 57% of the initial value, which indicating that most of the cake layer is removed. Flux values after second and third cleaning pulses are 1970 $L.h^{-1}.m^{-2}$ and 1965 $L.h^{-1}.m^{-2}$ at 185 and 235 minutes, respectively (56% and 55% of the initial value), the reproducibility is shown by error bars based on two experiments(Figure 5.12). When the second cleaning pulse which used SES (Figure 5.13) the pure water flux after the first, second and third cleaning pulses are 2292 $L.h^{-1}.m^{-2}$, 2220 $L.h^{-1}.m^{-2}$ and 1850 $L.h^{-1}.m^{-2}$ at 135, 185 and 235 minutes, respectively

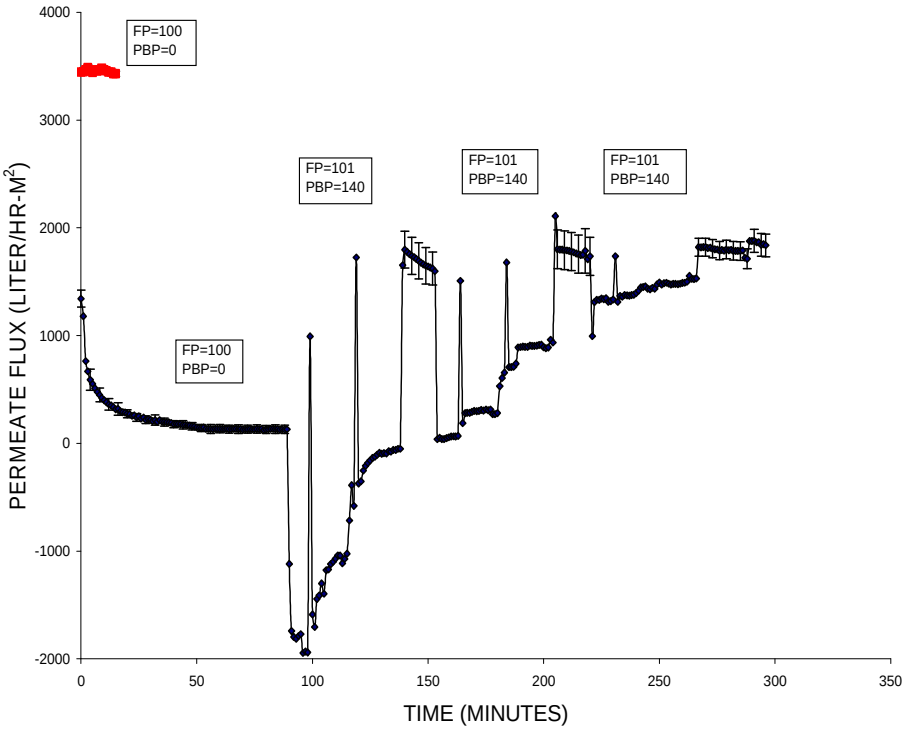


Figure 5.12 Flux against time for the Biodyne A (amphoteric nylon 6, 6) 0.45 μm membrane /yeast system (backpulsing with pure water). (FP: feed pressure, PBP: peak of backpulse pressure).

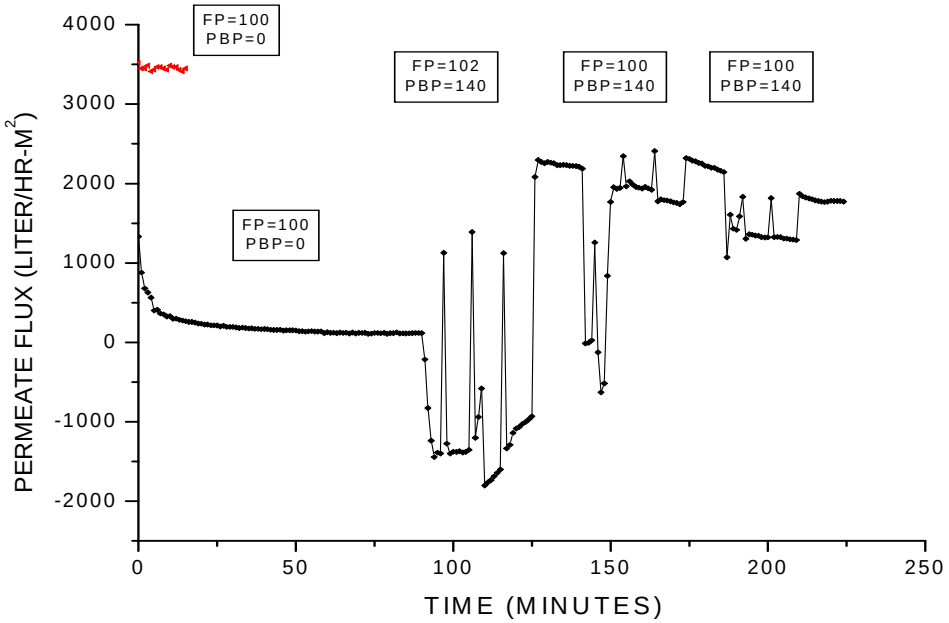


Figure 5.13 Flux against time for the Biodyne A (amphoteric nylon 6, 6) 0.45 μm membrane /yeast system (backpulsing with soap solution). (FP: feed pressure, PBP: peak of backpulse pressure).

(67, 64 and 53% of the initial value). The flux values after the third cleaning pulse using the soap solution, are clearly lower than after the third pure water cleaning pulse. The comparison of the flux values after cleaning by backpulsing for experiments with three pure water cleaning pulses and the second case where the second cleaning pulse was SES solution, this becomes evident (a possible reason can be, that the soap could lyse the yeast cells to create debris which may refoul the membrane during the periods where backpulsing is turned off in order to obtain flux measurements).

5.3.2 ULTRASONIC MEASUREMENTS

Figure 5.14 shows the changing amplitude of the reflected ultrasonic pulse recorded as a function of time at certain arrival times for the results given in Figure 5.12. The 0 minute signal (lower part of Figure 5.14) shows the peak generated from the pure water/new membrane interface and internal reflections from the membrane structure (Note that the structure of this membrane is similar to 0.2 µm nylon membrane). The density of the water-saturated upper layer of the nylon membrane would be very similar to density of water. So, the first peak on 0 minute signal is likely to have resulted from the central nylon support of the new membrane, before the fouling began there is a very small change in density from water to membrane. The arrival time for this peak was $5.3E^{-06}$ seconds. After filtration of the washed yeast suspension began, it was observed that, after 2 minutes of fouling, a peak due to the yeast layer becomes visible in front of the membrane peak. This peak is shifted towards earlier arrival times, up until 60 minutes, where the membrane is almost completely fouled. This is because of the gradual increase in the density and the thickness of the cake layer. The first peak on the 85 minute signal shows that a fouling layer still covered the membrane surface. The disappearance of the fouling peak after first cleaning pulse indicates that the backpulsing removes almost all of the caking fouling layer. The amplitude of the water/membrane peak on the 295 minute signal shows that the membrane properties (i.e. density) had changed compared with water/membrane peak at the 0 minute signal. This suggests that there are still some yeast cells that remain on the membrane surface and yeast cell debris inside the membrane pores (see Figure 5.15). The SEM image of the membrane cleaned by backpulsing proves the above observation (Figure 5.17(c)).

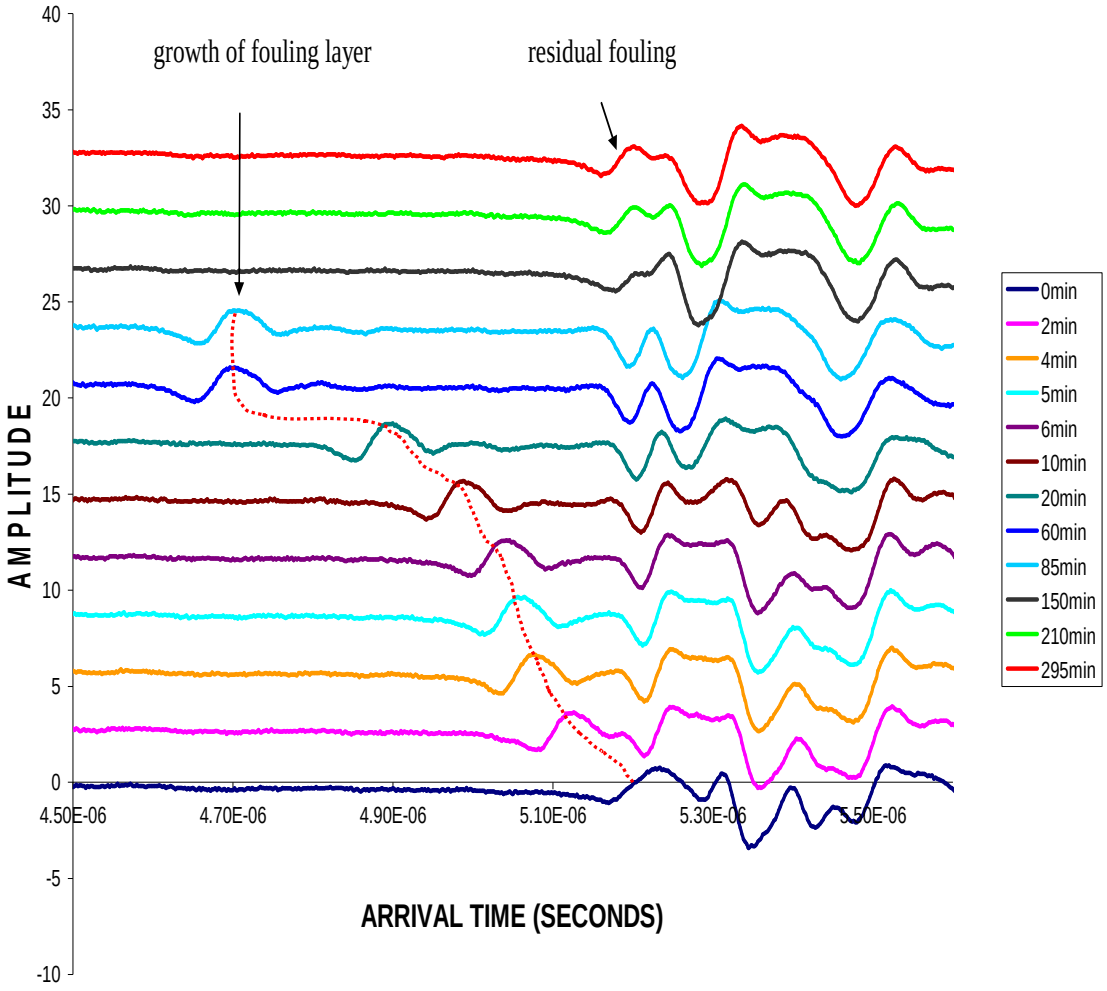


Figure 5.14 Amplitude of the reflection received at the detector as a function time, for the Biodyne A (amphoteric nylon 6, 6) 0.45 μm membrane/yeast system. The time interval shown encompasses all the reflections received for the water/film, film/membrane and membrane/metal support interfaces.

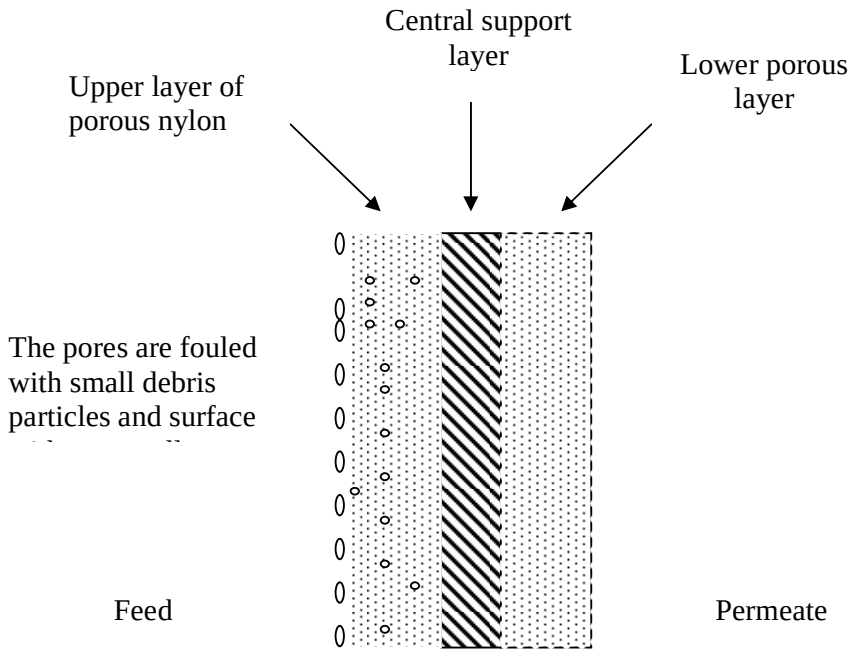


Figure 5.15 Proposed cross-section of the 0.45 µm nylon membrane cleaned by backpulsing (at 235 minutes).

5.3.3 DETERMINATION OF THE FOULING LAYER THICKNESS AS A FUNCTION OF TIME

As mentioned earlier the value of the fouling layer thickness was calculated each time using the time-distance relation equation (3.1). Figure 5.16 shows the fouling layer thickness as a function of time.

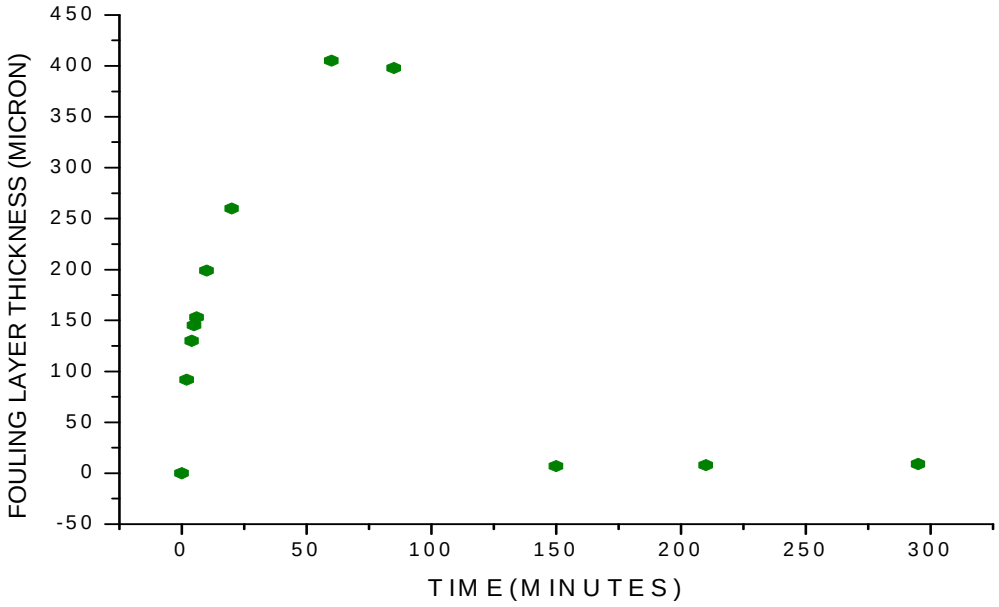
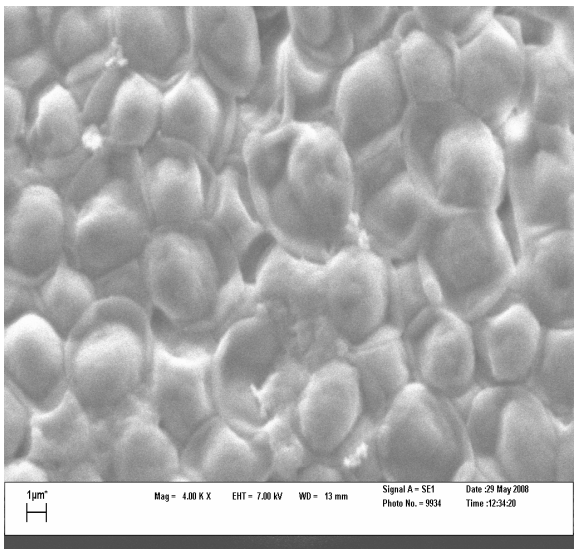


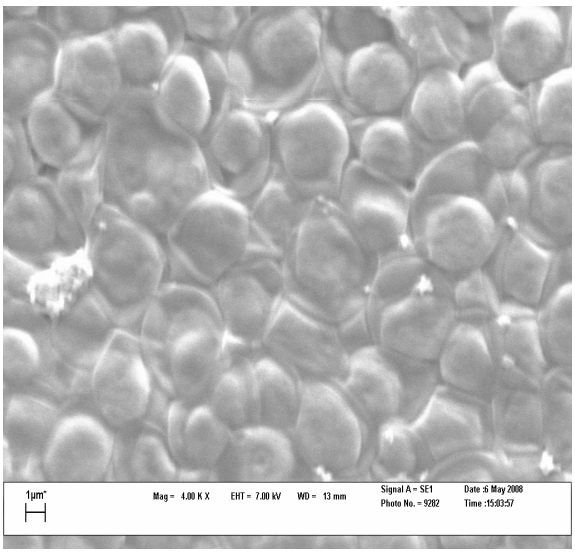
Figure 5.16 The fouling layer thickness for the Biodyne A (amphoteric nylon 6, 6) 0.45 µm membrane /yeast system as a function of time.

5.3.4 SEM ANALYSES

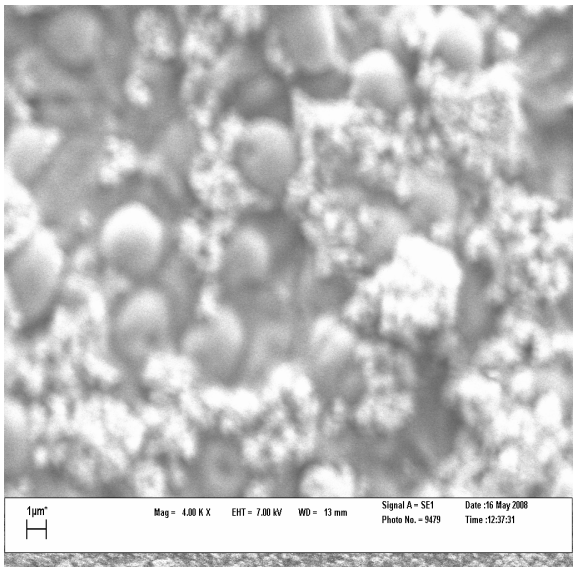
The new nylon 0.45 µm membrane surface structure is shown in Figure 4.8 (b). Figure 5.17 (a) shows the image of a fouled membrane after 15 seconds of normal cross-flow filtration. The yeast cells blocked or plug most of the membrane pores, causing the initial rapid drop in flux. Figure 5.17 (b) shows the image of the fouled membrane, after 60 minutes. As expected, there are now many more yeast cells on the membrane surface than the image after 15 seconds of filtration, so that now the membrane is completely covered by yeast cake layer, where the yeast cake layer thickness is 405 µm (see figure 5.16). An image of the cleaned membrane, taken after three pure water backpulses, is shown in Figure 5.17 (c).From this it can be seen that partial membrane cleaning is obtained by backpulsing; yeast cells are removed from some sections of the membrane surface while other sections remain covered and still blocks the membrane lowring the flux (The flux is about 65% that of the pure water flux). Figure 5.17 (d) shows an image of a cleaned membrane (when the second cleaning backpulse used soap solution). Some of yeast cells remain on the membrane surface and yeast debris is seen trapped on the pores and probably also in the pores of the membrane, furthermore, the cleaned membrane image (Figure 5.17 (d)) shows less porosity than the cleaned membrane (Figure 5.17 (c)). The flux is about 53% that of the pure water flux. High magnification SEM is representative of the surface as indicated by lower magnification images (see Appendix 2).



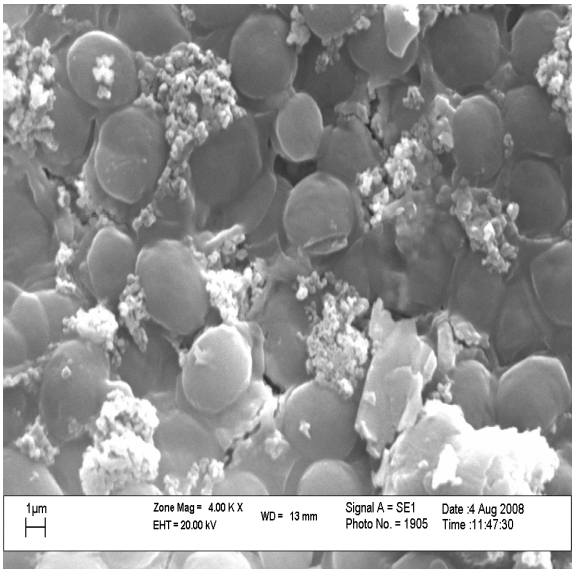
(a)



(b)



(c)



(d)

Figure 5.17 SEM images (magnification 4000X) of the Biodyne A (amphoteric nylon 6, 6) 0.45 µm membrane /yeast system. (a) membrane surface after being fouled for 15 seconds, (b) a fully fouled (60 minute) surface, (c) a surface cleaned by three pure water backpulses, (d) a surface cleaned with pure water, soap solution and pure water backpulses.

5.4 FOULING WITH A YEAST SUSPENSION IN A UF SYSTEM

5.4.1 DEFOULING EXPERIMENTS USING A THREE 140 KPA PULSE SEQUENCE

All experiments were carried out using a 100,000 MWCO PS membrane with a washed yeast suspension as a foulant during the cross-flow UF process. The test conditions for experiments are listed in Table 5.3.

Table 5.3 Summary of test conditions for yeast defouling experiment with PS membrane

Parameter	Value
Feed flowrate	0.045 ± .003L/min
Feed pressure	100 ±3 kPa
Temperature	25 ± 1◦C
Feed concentration	1 g/l
pH	8

Figures 5.18 and 5.19 are plots of the forward and reverse filtration flux as a function of time for washed yeast defouling experiments with three cleaning cycles, backpulsing with pure water during the first, second and third cleaning backpulses (Figure 5.18), and with pure water during the first, SES solution during the second and pure water during the third cleaning backpulses (Figure 5.19). In all cases the permeate flux decreases rapidly for the first 5 minutes, and slowly at longer time up to 60 minutes. For both experiments the fouled values at 60 minutes are about 70 $L.h^{-1}.m^{-2}$, which is 33% of the initial value (215 $L.h^{-1}.m^{-2}$), while after the pure water wash, the flux is about 85 $L.h^{-1}.m^{-2}$ (40% of the initial value). The following pure water flux values were recorded (Figure 5.18) after first, second and third cleaning backpulses using pure water and the average value is about 180 $L.h^{-1}.m^{-2}$ (90% of the initial value). When the second cleaning backpulse included use of SES solution (Figure 5.19), the same behaviour is observed, except that the flux values after the third cleaning backpulses are clearly lower than for all the pure water cleaning backpulses. According to the comparison of the flux values after cleaning by backpulsing for experiments with three pure

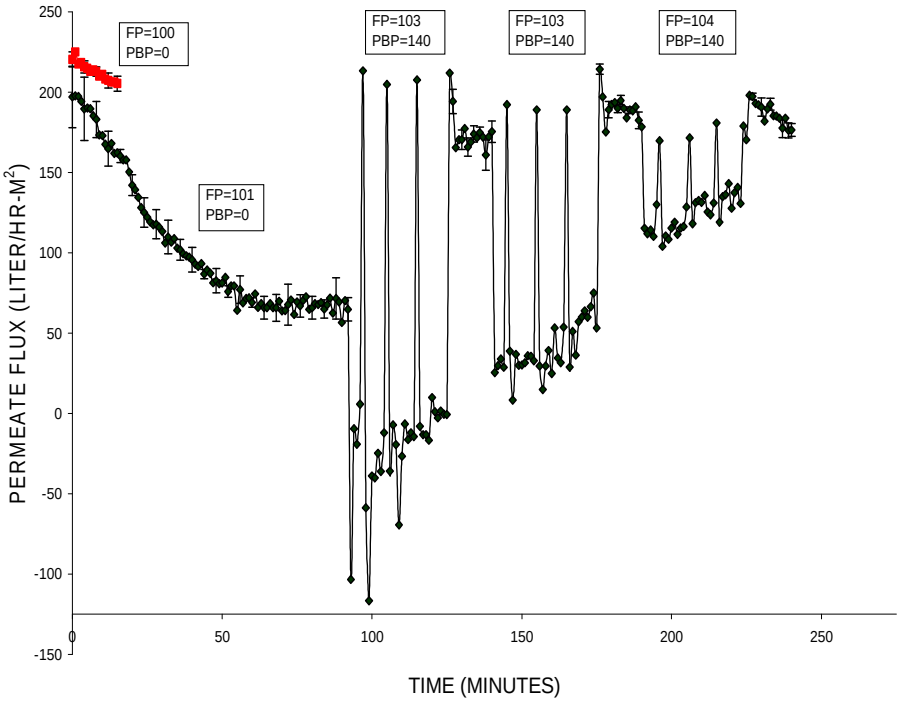


Figure 5.18 Flux against time for the PS membrane /yeast system (backpulsing with pure water). (FP: feed pressure, PBP: peak of backpulse pressure).

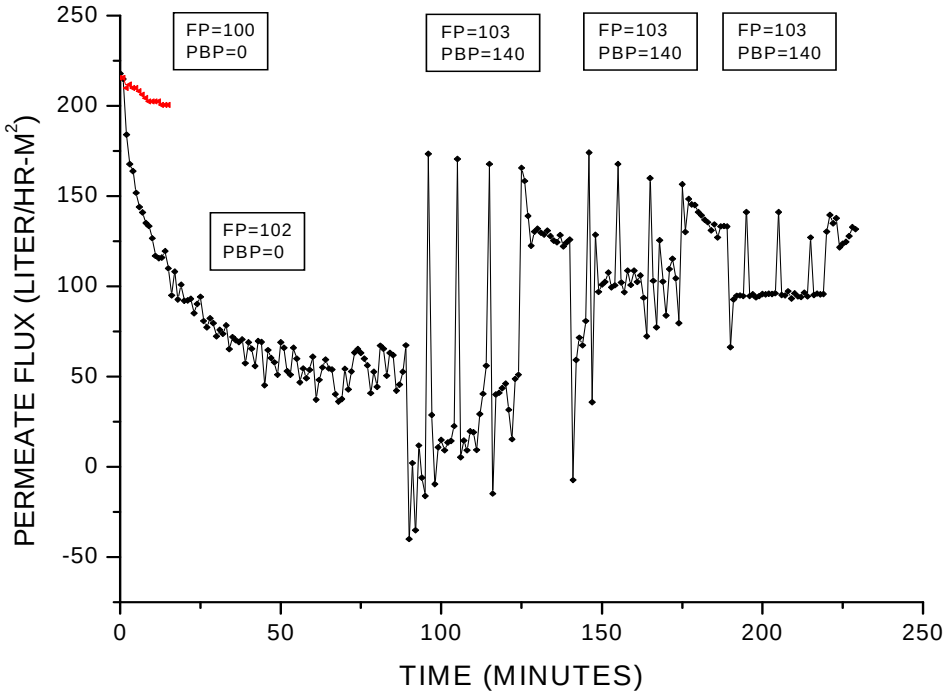


Figure 5.19 Flux against time for the PS membrane /yeast system (backpulsing with soap solution). (FP: feed pressure, PBP: peak of backpulse pressure).

water cleaning backpulses and when the second cleaning backpulse was SES solution, it can be concluded that the backpulsing cleaning with soap solutions does not improve the cleaning which decreases during the soap backpulsing.This could possibly be attributed to the soap lysing the yeast cells and refouling the membrane during periods of flux measurements.

5.4.2 ULTRASONIC MEASUREMENTS

Figure 5.20 shows the changing amplitude of the reflected pulse recorded as a function of certain arrival times for the results given in Figure 5.18. The 0 minute signal (lower trace of Figure 5.20) shows the peak generated from the pure water/new membrane interface and internal reflections from the membrane structure. The arrival time for this peak was 5.23 E^{-06}seconds. After filtration of the washed yeast suspension began, as can be seen the new peak started, to build up in front of the membrane peak which is due to formation of yeast layer. This is observed up to 60 minutes, where the membrane reached a near steady-state flux. The fouling peaks, at the early stages of operation time, cannot be seen clearly. This is because of the formation of a very thin cake layer on the membrane surface compared with the PS membrane layer. The fouling peak was almost completely separated and defined after 20 minutes of operation time. The results showed that the thickness of the fouling layer is 199 µm (see section 5.4.3), the cleaned membrane signal reflection at 135, 185 and 235 minutes shows that there is a peak visible in front of the membrane peak, and this peak could be due to some yeast cells still on the cleaned membrane surface (see Figure 5.21). This is supported by the remaining fouling layer thickness (Figure 5.22).

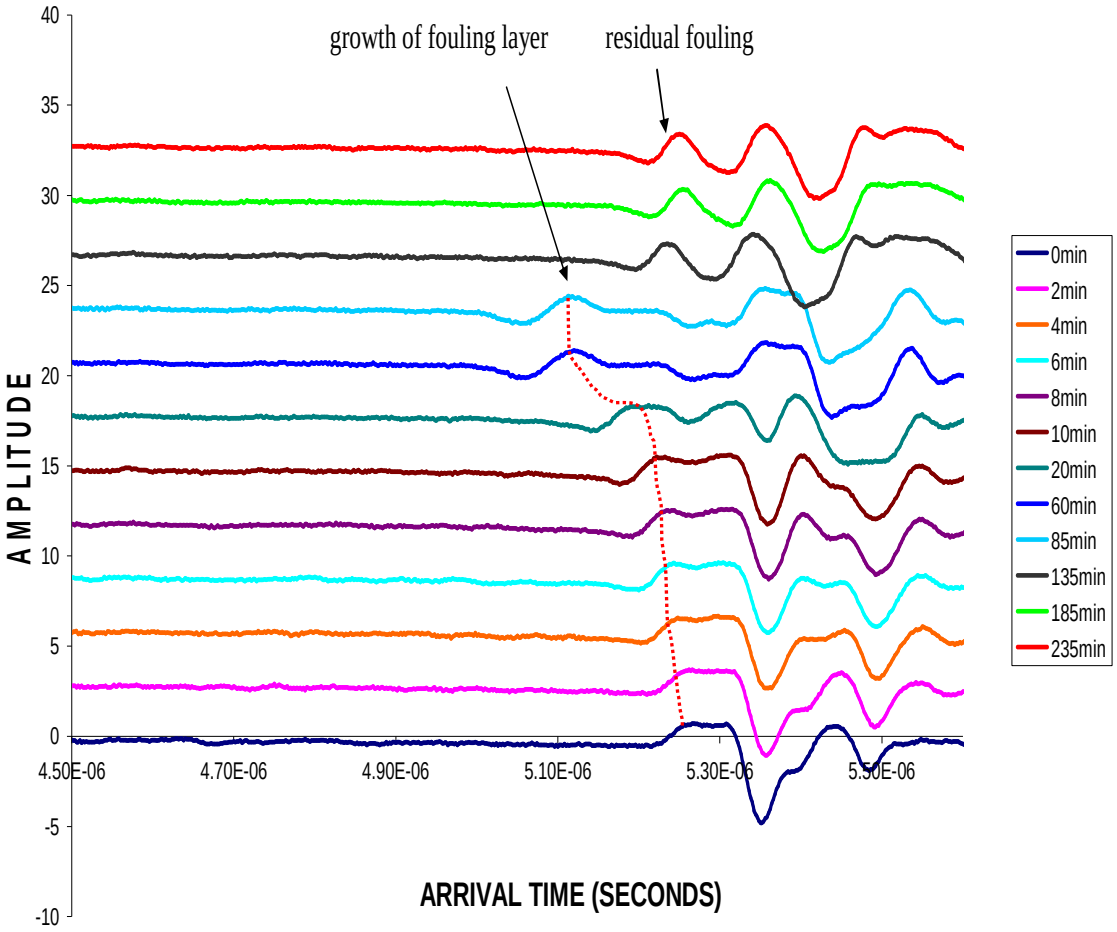


Figure 5.20 Amplitude of the reflection received at the detector as a function time, for the PS membrane/yeast system. The time interval shown encompasses all the reflections received for the water/film, film/membrane and membrane/metal support interfaces.

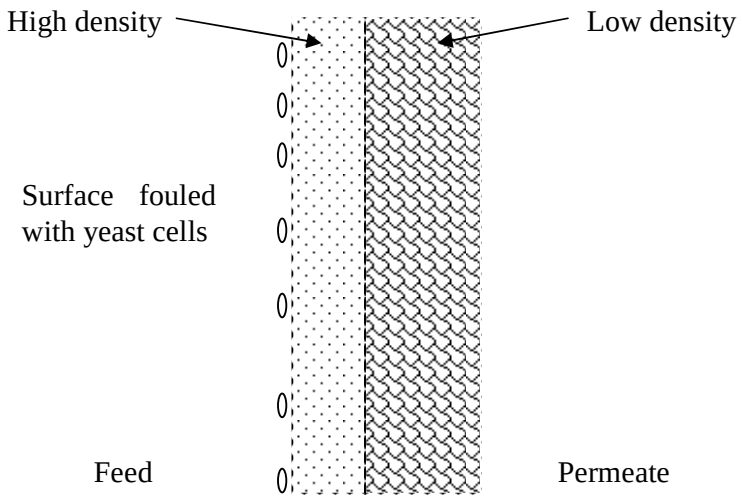


Figure 5.21 Proposed cross-section of the PS membrane cleaned by backpulsing (at 235 minutes).

5.4.3 DETERMINATION OF THE FOULING LAYER THICKNESS AS A FUNCTION OF TIME

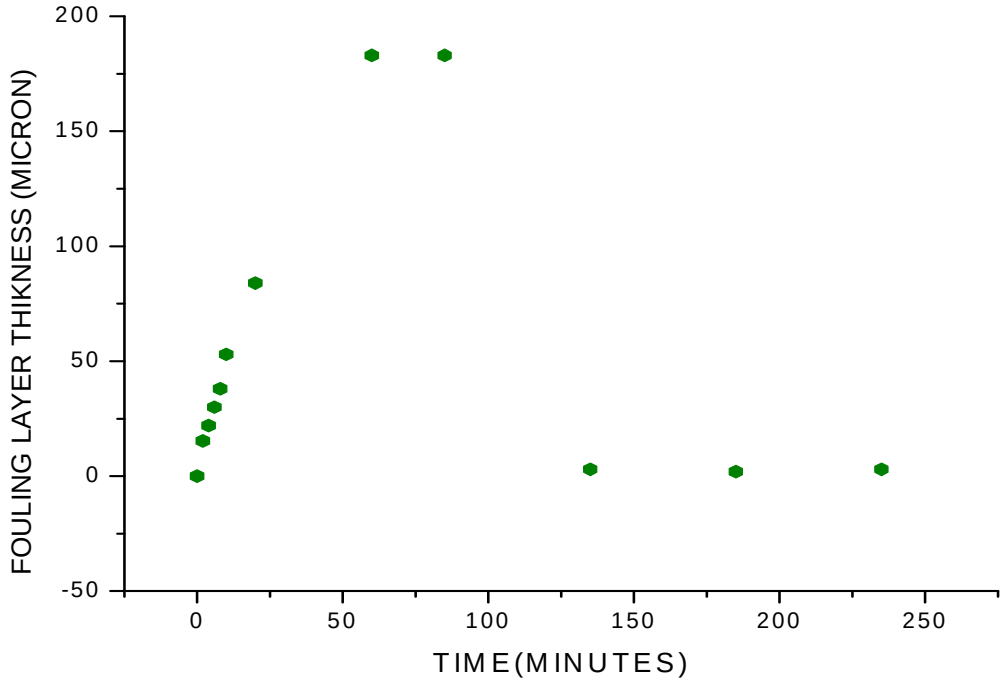
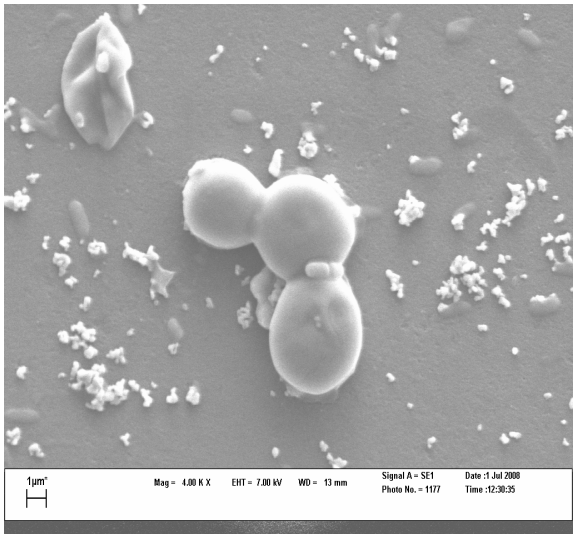


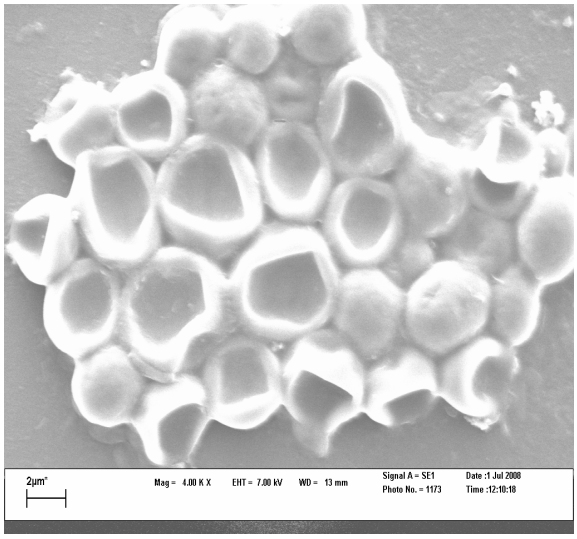
Figure 5.22 The fouling layer thickness for the PS membrane /yeast system as function of time.

5.4.4 SEM ANALYSES

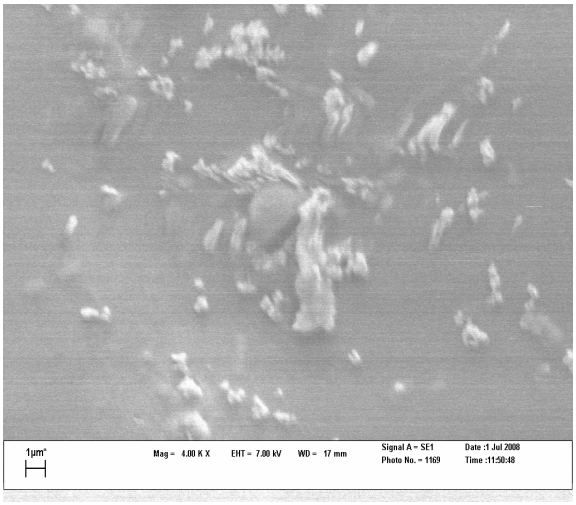
A new 100, 000 MWCO PS membrane surface structure is shown in Figure 4.9, Figure 5.23(a) shows the image of a fouled membrane for normal cross-flow filtration after 15 seconds. The image shows some yeast cells on the membrane surface. The fouled membrane image, taken after 60 minutes, given in Figure 5.23 (b) shows more deposited yeast cells which are covering the membrane surface. The cleaned membrane image (with three pure water backpulses) is shown in Figure 5.23 (c), taken after the third cleaning pulse. It shows that, almost complete membrane cleaning is obtained by backpulsing, and almost all of the yeast layer is removed. But there is still some yeast cell debris that remains on the membrane surface after backpulsing cleaning. Figure 5.23 (d) shows the image of a cleaned membrane (where the second cleaning backpulse was soap solution), the yeast cells were completely removed from some patches of the membrane surface, while other patches were still covered by yeast cells and yeast cell debris. High magnification SEM is representative of the surface as indicated by lower magnification images (see Appendix 2).



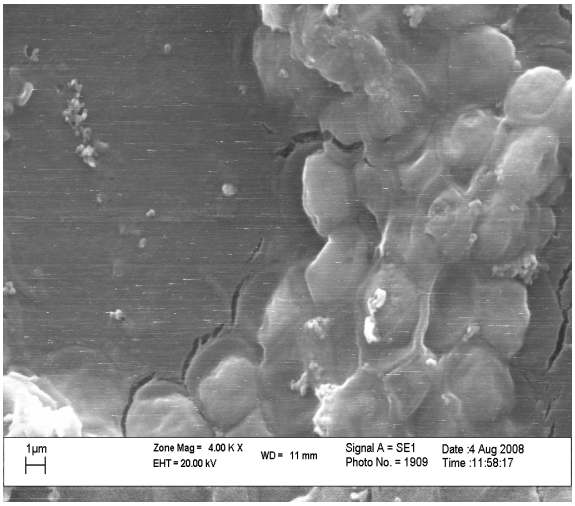
(a)



(b)



(c)



(d)

Figure 5.23 SEM images (magnification 4000X) of the PS membrane /yeast systems. (a) membrane surface after being fouled for 15 seconds, (b) a fully fouled (60 minute) surface, (c) a surface cleaned by three pure water backpulses, (d) a surface cleaned with pure water, soap solution and pure water backpulses.

5.5 FOULING WITH A DEXTRIN SOLUTION IN A UF SYSTEM

5.5.1 DEFOULING EXPERIMENTS USING A THREE 140 KPA PULSE SEQUENCE

All experiments were carried out using a 100,000 MWCO PS membrane with dextrin as a foulant during the cross-flow UF. The test conditions used for the experiments are listed in Table 5.4.

Table 5.4 Summary of test conditions used for the dextrin defouling experiment with PS membrane

Parameter	Value
Feed flowrate	0.045 ± .003L/min
Feed pressure	100 ±3 kPa
Temperature	25 ± 1◦C
Feed concentration	0.5 g/l
pH	7.5

Figures 5.24 and 5.25 are plots of the forward filtration flux as a function of time for dextrin solution defouling experiments with three cleaning cycles: backpulsing with pure water during the first, second and third cleaning backpulses (Figure 5.24), and with pure water during the first, with SES solution during the second and with pure water during the third cleaning pulses (Figure 5.25). In all cases the permeate flux decreases rapidly at first and then at a decreased rate up to 60 minutes, where the flux values were about 26% of the initial value. In both of the figures, after the pure water wash, the flux values were about 32%. In both cases the first cleaning backpulsing increased the water flux values to about 85% of their initial values, the second cleaning backpulsing increase the flux values by a small amount to about 97% of their initial values, after the third cleaning backpulsing the flux values are above the initial pure water values (> 100%). These results show that, there is no need for soap solutions.

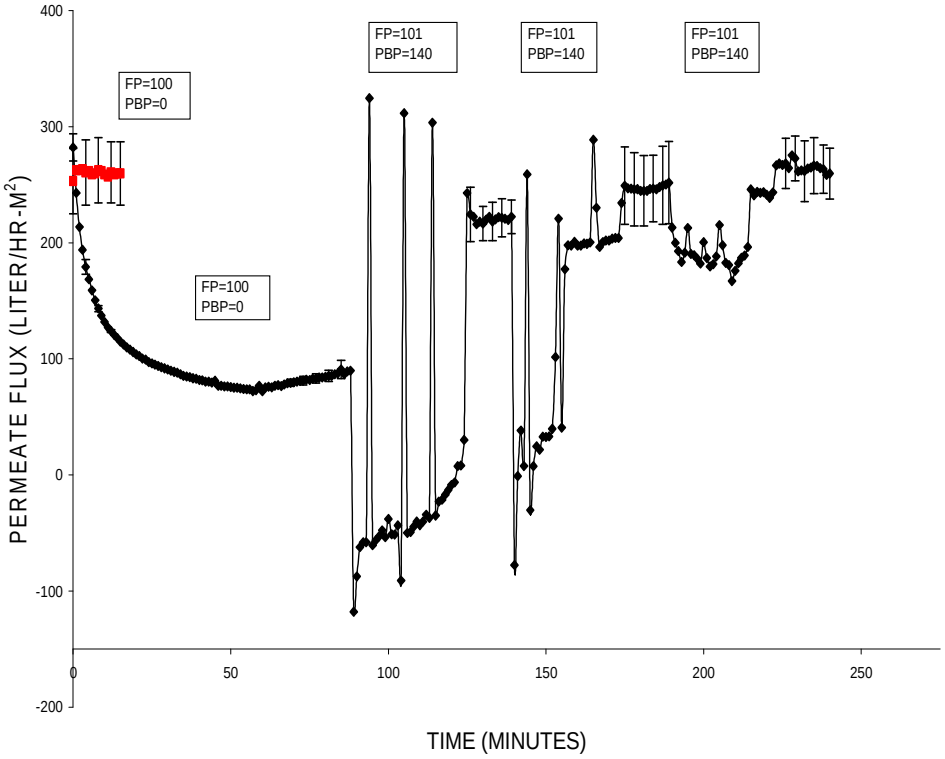


Figure 5.24 Flux against time for the PS membrane /dextrin system (backpulsing with pure water). (FP: feed pressure, PBP: peak of backpulse pressure).

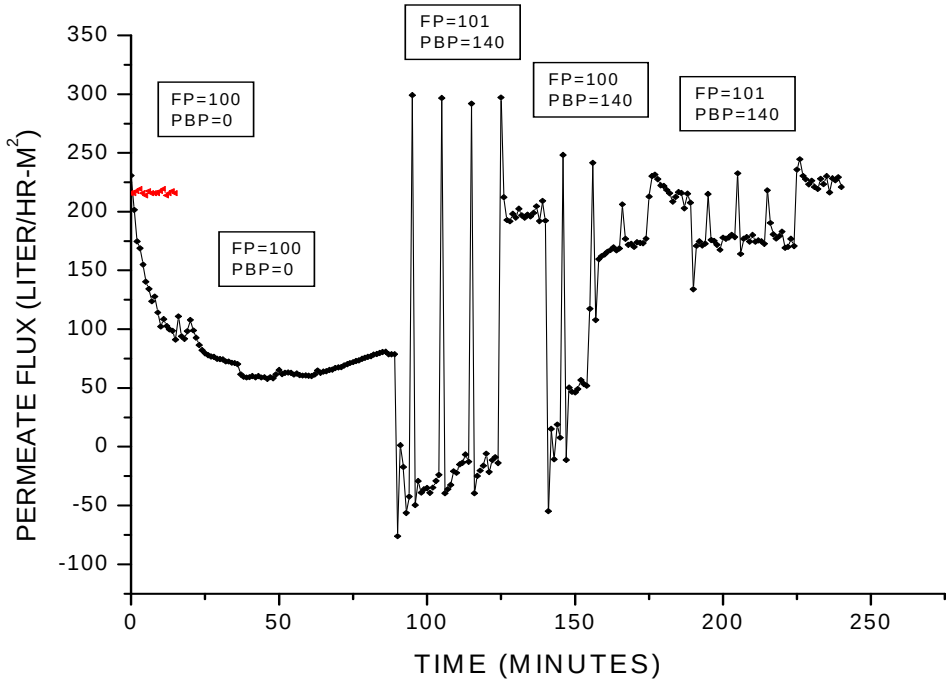


Figure 5.25 Flux against time for the PS membrane /dextrin system (backpulsing with soap solution). (FP: feed pressure, PBP: peak of backpulse pressure).

5.5.2 ULTRASONIC MEASUREMENTS

Figure 5.26 shows the changing amplitude of the reflected pulse recorded as a function of time at certain arrival times for the results given in Figure 5.24. The 0 minute signal shows the peak generated from the pure water/new membrane interface and internal reflections from the membrane structure. The arrival time for this peak was $5.23E^{-06}$seconds. After filtration of the dextrin solution began, some of dextrin particles began to be adsorbed onto the membrane surface and into the pores of the membrane. There is a continual decrease in permeate flux visible from the beginning (see Figure 5.24), indicating the initiation of fouling due to dextrin molecular adsorption. The dextrin adsorption would lead to small changes in the amplitude of the water/ membrane peak because of the changes in membrane properties (i.e. density).The water/fouling peak cannot been seen during the operating time of the experiment. This is because the dextrin forms a very thin cake layer (compared with the PS membrane layer) with a density very similar to the density of water. After cleaning by backpulsing, the amplitude of the water/membrane peaks changes and all that can be seen is a compaction and membrane density changes (see Figure 5.27). In the case of a very thin cake layer, it is difficult to calculate the thickness of the fouling layer using equation 3.1. Because of the echo signal of the cake layer is combined with the echo signal of the PS membrane layer, so it is difficult to know which the real echo of the cake layer is.

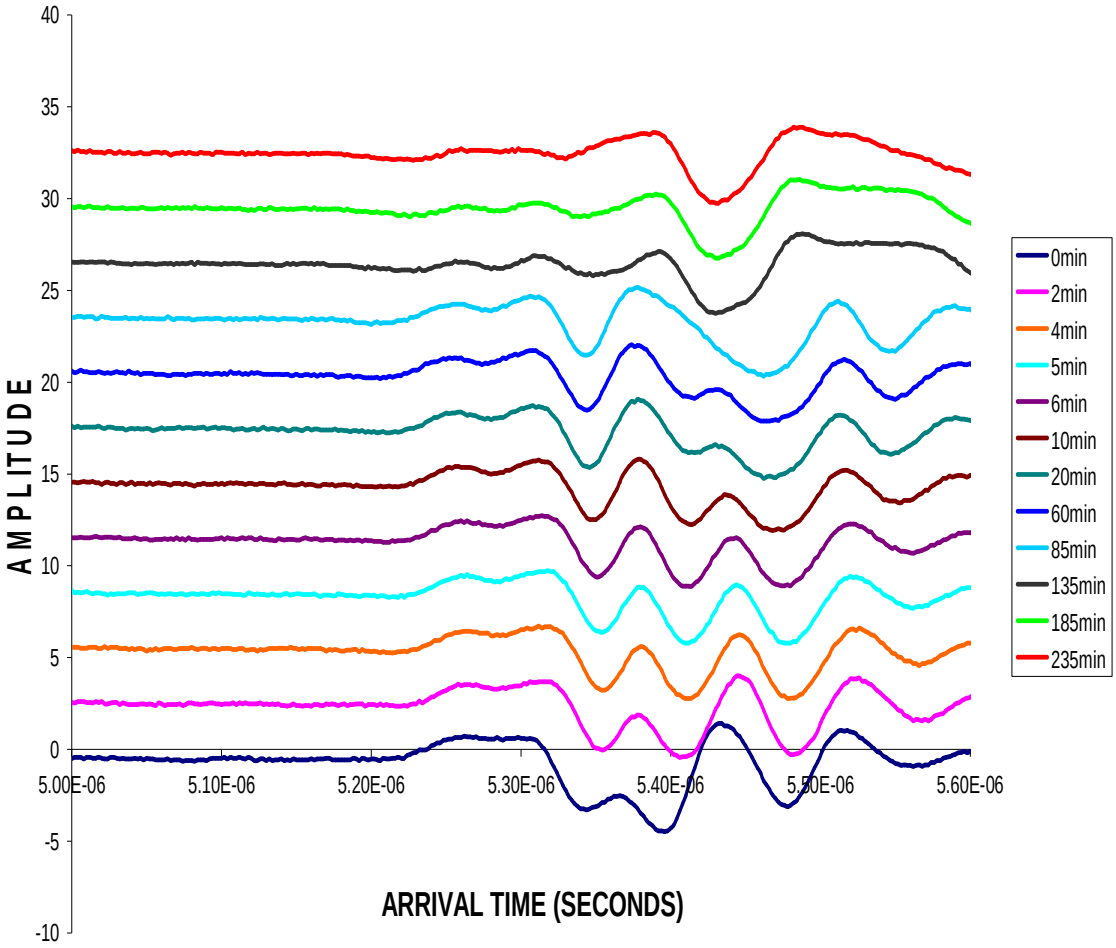


Figure 5.26 Amplitude of the reflection received at the detector as a function time, for the PS membrane/dextrin system. The time interval shown encompasses all the reflections received for the water/film, film/membrane and membrane/metal support interfaces.

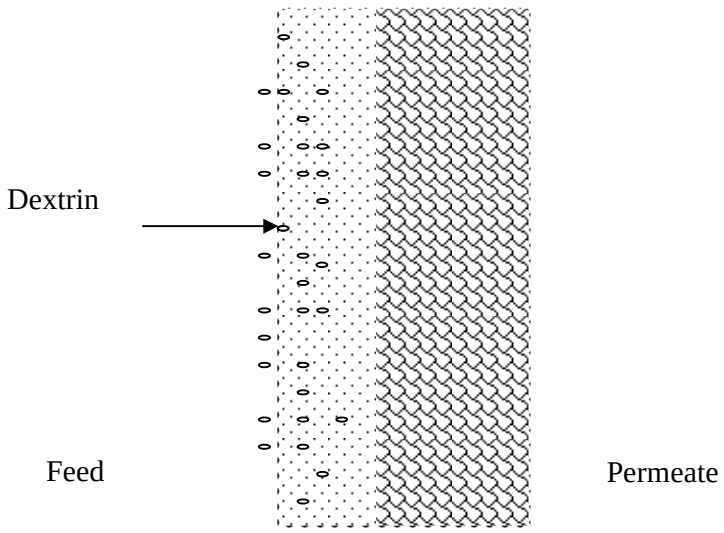
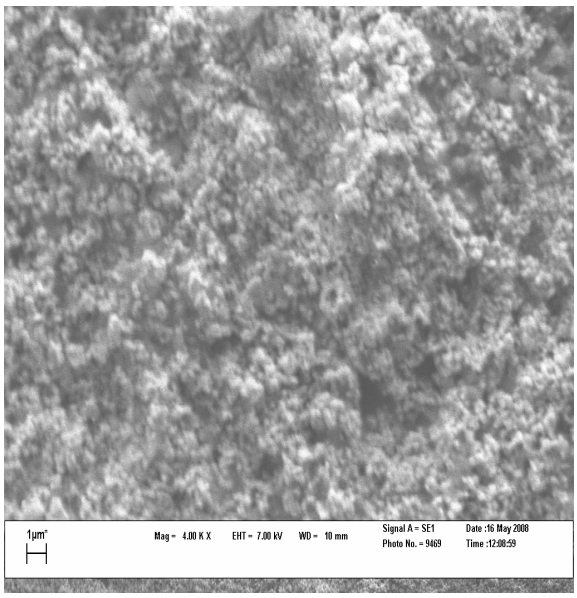


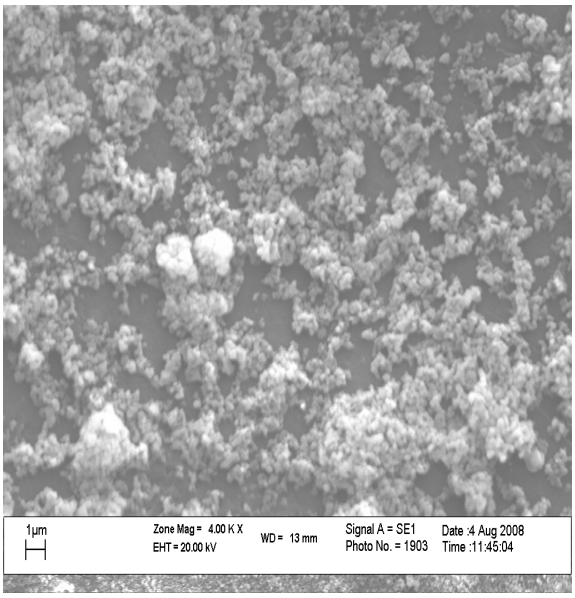
Figure 5.27 Proposed cross-section of the PS membrane cleaned by backpulsing (at 235 minutes).

5.5.3 SEM ANALYSES

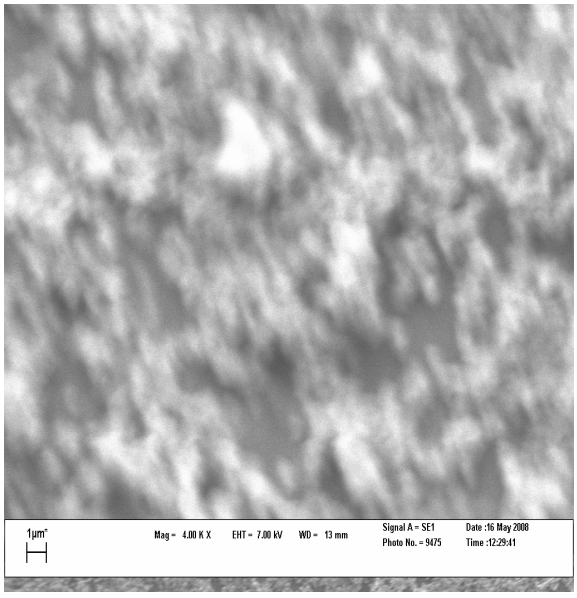
The fouled membrane image (new membrane image given in Figure 4.9) given in Figure 5.28 (a), taken after 60 minutes, shows that there is an amount of dextrin agglomerates accumulated on the membrane surface, and the ultrasonic results given in Figure 5.26, shows no clear fouling peak in front of the membrane peak. This would mean that the layer is very thin or more plausibly that the dextrin layer has the same density as water. The cleaned membrane (with three pure water backpulses) is shown in the image in Figure 5.28 (b), taken after the third cleaning backpulse. Some of the particles have been removed by backpulsing but many remain. Here the flux was above the initial pure water flux values. Figure 5.28 (c) and (d) show images of the cleaned membrane (when the second cleaning backpulse involved the use of soap solution). High magnification SEM is representative of the surface as indicated by lower magnification images (see Appendix 2).



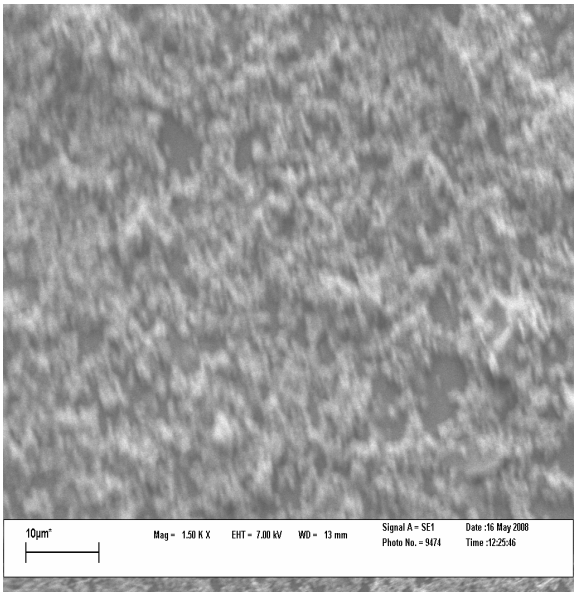
(a)



(b)



(c)



(d)

Figure 5.28 SEM images (magnification 4000X) of the PS membrane /dextrin system. (a) Membrane surface after being fouled for 60 minute, (b) a surface cleaned by three pure water backpulses, (c) and (d) a surface cleaned by pure water, soap solution and pure water backpulses.

5.6 DISCUSSION

Representative experimental results are summarized in the tables given below. The membranes used in the experiments are, a PAL Biodyne 0.2 nylon membranes (P2 in the tables), a PAL Biodyne 0.45 nylon membranes (P1 in the tables) and Alpha Leval GRO 100,000 MWCO polysulfone membranes (G in the tables). Foulants used are, alumina (A in the tables), washed yeast (Y in the tables) and dextrin (D in the tables). The soap solutions used are SES soap solution (S in the tables) and F9 soap solution (F in the tables). Note that some results of experiments using F9 soap solution are illustrated in Appendix 2. Pure water is R in the tables. In the tables the first column is M/F (membrane/ foulant) and the second column is RO (pure water flux values). The other columns are 5 min (flux value after fouling for 5 minutes), 60 min (flux value after fouling for 60 minutes), 90 min (flux value after washing), BP1 (flux values after first backpulsing cleaning cycle), BP2 (flux values after second backpulsing cleaning cycle), BP3 (flux values after third backpulsing cleaning cycle).

EXPERIMENTS USING A 90 KPA, 140 KPA, 180 KPA SEQUENCE

As previously mentioned these experiments were carried out using three backpulsing cleaning cycles, namely 90 kPa, 140 kPa and 180 kPa peak backpulse pressures. In the early experiments, some of the experiments were stopped after the second backpulsing cleaning cycle (140 kPa). The third backpulsing cleaning cycle (180 kPa) was used only after the flat-cell membrane module had been modified to have a spacer cloth above the membrane to prevent serious bulging of the membrane. The results of these experiments are summarized in Table 5.5 and are illustrated in Figures 5.1, 5.2 and 5.3. The results in Table 5.5 indicate that almost no difference between the three pure water backpulsing results and backpulsing results with a soap solution.

EXPERIMENTS USING A THREE 140 KPA PULSE SEQUENCE

These experiments were carried out using three backpulsing cleaning cycles at 140 kPa peak backpulse pressure. Representative experimental results are illustrated in Figures 5.4, 5.5, 5.12, 5.13, 5.18, 5.19, 5.24, and 5.25 and summarized in Table 5.6.

For the two nylon membranes (P1, P2), it can be seen that the flux decreased rapidly in the first 15 seconds of operation. This is because of the blocking of the pores of the membranes by the foulant particles. This is followed by a gradual decrease of permeate flux over the next

60 minutes of operation. For polysulfone membranes the flux also decreases rapidly at first and then more slowly (i.e. less than that of the nylon membranes) after longer periods up to 60 minutes.

Table 5.5 Representative experimental results of experiments using a 90 kPa, 140 kPa, and 180 kPa sequence.

NO	M/F	RO ($L.h^{-1}.m^{-2}$)	5MIN ($L.h^{-1}.m^{-2}$)	60MIN ($L.h^{-1}.m^{-2}$)	90MIN ($L.h^{-1}.m^{-2}$)	BP1 ($L.h^{-1}.m^{-2}$)	BP2 ($L.h^{-1}.m^{-2}$)	BP3 ($L.h^{-1}.m^{-2}$)
1	P2/A-R	4699	1312.5	337.5	375	450	4658	4668
2	P2/Y-R	1818.75	1500	58.3	58.2	266	343	396
3	P2/Y-S	2625	1687.5	86	26.6	22	24	89.5
4	P1/Y-F	937.5	562.5	68	93.75	82.5	349	231
5	P1/Y-R	3720	431	129.5	127.5	131	2495	2650
6	G/Y-R	289	187.5	92	81	81	187.5	
7	G/Y-R	113	94	32	48	70	90	100
8	G/D-R	190.5	166.5	47	56	71.5	88	
9	G/D-R	223	172.5	47	52.5	112.5	101	
10	G/D-R	157.5	131	50.5	70	101	101	122
11	G/D-S	164	137	49	37.5	39	97.5	
12	G/D-S	176	154	47	34.5	43	84	
13	G/D-F	172.5	159	49	22.5	49	145	
14	G/D-F	197	152	54	28	79	79	169

The membranes were then washed for 30 minutes using pure water. In all cases only small changes were observed in the flux values before and after washing (between 60 and 90 minutes). The membrane was then backpulsed three times. Recall that the first backpulsing cleaning cycle was carried out using pure water; the second backpulsing cleaning cycle was carried out using pure water or a soap solution and the third backpulsing cleaning cycle was carried out using pure water.

Table 5.6 (1 – 6) shows representative results when an alumina suspension is used with the Biodyne A (amphoteric nylon 6, 6) 0.2 μm MF membrane (P2), which are illustrated in Figures 5.4 and 5.5. The table shows these flux values at 60 minutes (fouled values) lay between 15% and 20% of the initial pure water flux value, while at 90 minutes (washed values) the permeate flux value is almost the same as fouling permeate flux value. It indicates

that the pure water cannot, without backpulsing, effectively clean the membrane. The BP1, BP2 and BP3 flux values, with and without the soap solutions, lay between 90% and 98%. This is because the alumina powder has an average diameter of 1.0 µm, and should not enter or stick to the 0.2 µm pores of the membrane. The results show the pure water backpulsing is very effective in cleaning the membrane and there is no need for soap solutions. An indication of the reproducibility is given by Figure 5.5 (second backpulsing with soap solution), where the permeate flux curves are similar, despite the difference in the types and the concentrations of the solutions.

Table 5.6 (7 – 14) shows representative results when yeast suspension is used with the Biodyne A (amphoteric nylon 6, 6) 0.45 µm MF membrane (P1), which are illustrated in Figures 5.12 and 5.13. The table shows the flux values at 60 minutes (fouled values) which lay between 25% and 30% of the initial pure water flux value, while at 90 minutes (washed values) the flux values are somewhat lower. The yeast cells have a diameter much larger than the alumina, but are deformable, and the soft yeast cells very effectively block the membrane pores. This is supported by the SEM results in Section 5.3.4. In all cases the BP1, BP2 and BP3 flux values, with and without soap solutions, have a tendency to stay the same or decrease from BP1 to BP3, particularly, with soap solution experiments. This is because the soap could lyse the yeast cells to create debris which refouls the membrane during the periods for flux measurements. This is supported by SEM results (Figure 5.17 (d)). From Table 5.6 it can be seen that, the BP1 flux values show the major removal of the foulant during the first backpulsing cleaning cycle. This is supported by UTDR results (Figure 5.14). The results show there is no need for soap solutions. An indication of the reproducibility is given by Figure 5.13 (second backpulsing with soap solution), where the permeate flux curves are similar, despite the difference in the types and the concentrations of the solutions.

Table 5.6 (15 – 20) shows representative results when a washed yeast suspension was used with the PS UF membrane (G), which are illustrated in Figures 5.18 and 5.19. The table shows that the flux values at 60 minutes (fouled values) lay between 30% and 35% of the initial pure water flux value, while at 90 minutes (washed values) the flux values lay between 30% and 40%. For yeast, using pure water for all three backpulsing cleaning cycles, the BP1, BP2 and BP3 flux values stay together at 90%. Using the soap solutions the same behaviour is observed, except the BP3 flux values are clearly lower than for all the pure water cleaning pulses, which also tended to decrease from BP1 to BP2 (note that this also happened for yeast with the Biodyne A (amphoteric nylon 6, 6) 0.45 µm MF membrane using soap solutions). This negative slope could be characteristic of the yeast foulant. This is supported by SEM

Table 5.6 Representative experimental results of experiments using a three 140 kPa backpulse sequence.

NO	M/F	RO ($L.h^{-1}.m^{-2}$)	5MIN ($L.h^{-1}.m^{-2}$)	60MIN ($L.h^{-1}.m^{-2}$)	90MIN ($L.h^{-1}.m^{-2}$)	BP1 ($L.h^{-1}.m^{-2}$)	BP2 ($L.h^{-1}.m^{-2}$)	BP3 ($L.h^{-1}.m^{-2}$)
1	P2/A-R	2906	1181	375	384	2850	2812.5	1969
2	P2/A-R	2960	1594	544	544	2560	2650	2890
3	P2/A-R	2925	1162.5	225	412.5	3000	3131	3094
4	P2/A-S	3125	1500	394	328	3115	3110	3125
5	P2/A-S	3170	1237.5	450	450	3140	3145	3155
6	P2/A-F	2662.5	1406	469	469	3150	2962.5	3075
7	P1/Y-R	2812	675	172.5	165	2081	1012	618
8	P1/Y-R	3465	562.5	175	167	1990	1970	1965
9	P1/Y-R	3469	431	112	96	2438	1856	1312
10	P1/Y-S0.5	3506	450	103	88	3675	2625	750
11	P1/Y-S1	3450	562	126	116	2006	2212	1818
12	P1/Y-S1	3188	489	154	135	2292	2220	1850
13	P1/Y-S 2	3562	412	112	109	3694	1500	562
14	P1/Y-F	2907	563	156	169	3150	2100	1050
15	G/Y-R	230	193	64	64	188	201	207
16	G/Y-R	215	201	70	85	180	181	182
17	G/Y-S0.5	206	169	60	66	131	139	139
18	G/Y-S1	169	165	62	60	161	150	143
19	G/Y-S 2	221	201	66	56	128	120	109
20	G/Y-F	218	183	62	56	178	118	128
21	G/D-R	242	169	84	101	221	221	169
22	G/D-R	262	178	71	79	238	255	256
23	G/D-R	236	171	68	84	202	214	188
24	G/D-R	281	172	75	96	253	272	289
25	G/D-S	223	176	92	96	188	199	234
26	G/D-S	219	159	86	101	181	218	232
27	G/D-F	197	152	81	99	180	216	222
28	G/D-F	214	163	86	109	162	132	165

results (Figure 5.23 (d)). It can again be concluded that backpulsing with soap solutions does not improve the cleaning process. An indication of the reproducibility is given by Figure 5.19 (second backpulsing with soap solution), where the permeate flux curves are similar, despite the difference in the types and the concentrations of the solutions.

Table 5.6 (21 – 28) shows representative results when a dextrin solution was used with the PS UF membrane (G), which are illustrated in Figures 5.24 and 5.25. The table shows the flux values at 60 minutes (fouled values) which lay between 40% and 55% of the initial pure water flux value, while at 90 minutes (washed values) the flux values are lay between 60% and 65%. For dextrin, with and without soap solutions, the BP1, BP2 and BP3 flux values improved from BP1 to BP2. The change of the flux values from BP1 to BP3 gave both positive and negative results. From these results it can be concluded that when a dextrin solution was used with the PS UF membrane there is no need for soap solutions and excellent membrane regeneration is achieved.

5.7 EXPERIMENTS WITH COLLOIDAL FOULANTS

These experiments were done to check the effect of the colloidal particles which are associated with the alumina and the yeast on the fouling mechanism. Colloidal alumina suspension was accumulated from washing of 1 g of alumina and mixed with 5 liters of pure water. The yeast colloidal suspension was accumulated from 1 g of yeast and mixed with 5 liters of pure water.

When the colloidal alumina suspension was used with nylon 0.2 µm membrane (Figure 5.29), the results show that the flux values at 60 minutes and after the first, second and third cleaning backpulses are lower than those of the normal 1 g/liter of washed alumina. This shows that the colloidal suspension is a more effective foulant.

When the colloidal yeast suspension is used with the nylon 0.45 µm membrane (Figure 5.30), the flux values decrease from first cleaning backpulse to the second cleaning backpulse to the third cleaning backpulse. This decrease is due to blocking of the pores of the membrane by the colloidal particles. When the yeast colloidal suspension is used with PS membranes (Figure 5.31), the results show that the flux values were very similar to those of the normal washed yeast experiments but the flux values after the cleaning by backpulsing are worse than those of the normal washed yeast as fine cell debris enter and block pores.

From the above results it can be concluded that backpulsing is not effective in removing the colloidal particles from both nylon membranes, but is better than in the case of removing yeast colloids from PS membranes.

As a control blank experiments were carried out using a two 140 kPa backpulse sequence, these experiments were carried out using a soap solution and not pure water during the first cleaning of backpulsing to see if the first backpulse cycle give higher flux values after either the first or the second backpulsing cleaning cycle when compared to the previous three backpulsing cleaning cycles experiments. The results of these experiments are illustrated in Figures A4.1 – A4.4 (Appendix 4). In all cases the results show that the flux values after both first and second backpulsing cleaning cycles were very similar to those observed in the three backpulsing cleaning cycle experiments.

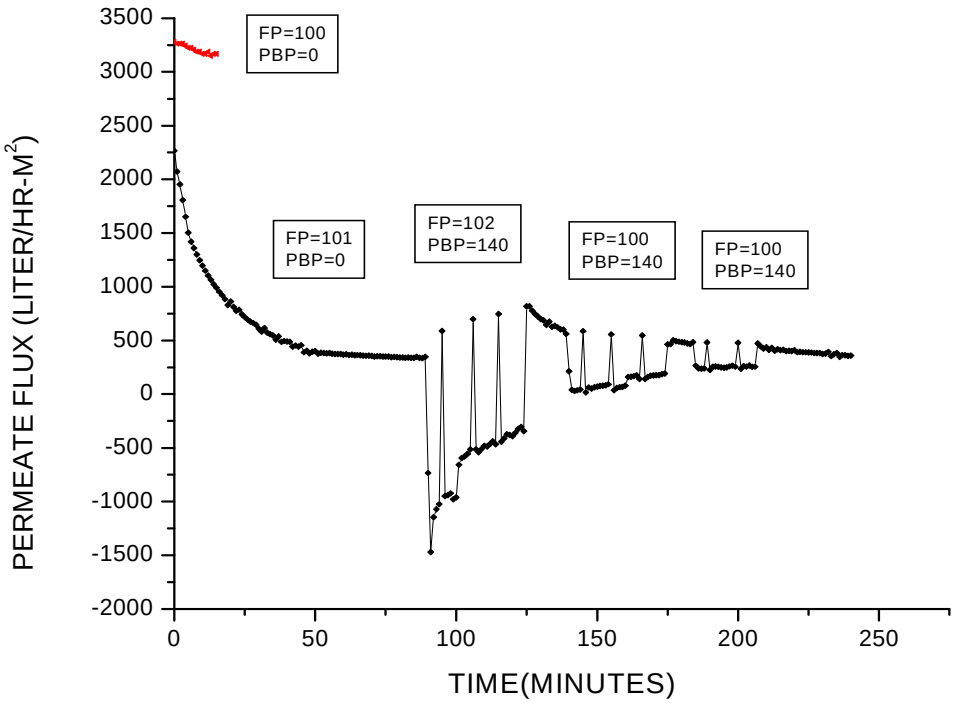


Figure 5.29 Flux against time for the Biodyne A (amphoteric nylon 6, 6) 0.2 μm membrane /alumina colloidal suspension system. (FP: feed pressure, PBP: peak of backpulse pressure).

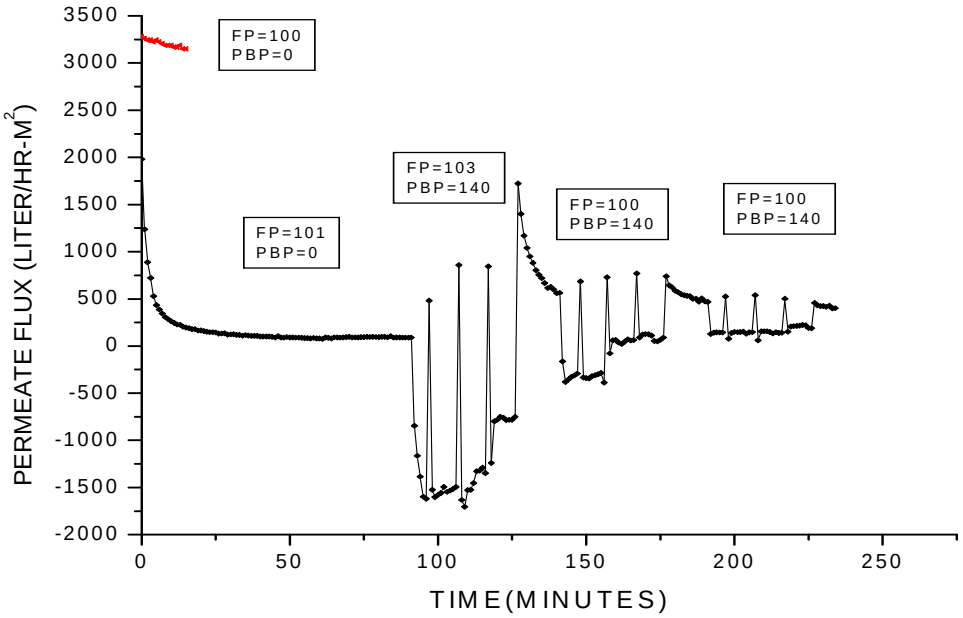


Figure 5.30 Flux against time for the Biodyne A (amphoteric nylon 6, 6) 0.45 μm membrane /yeast colloidal suspension system. (FP: feed pressure, PBP: peak of backpulse pressure).

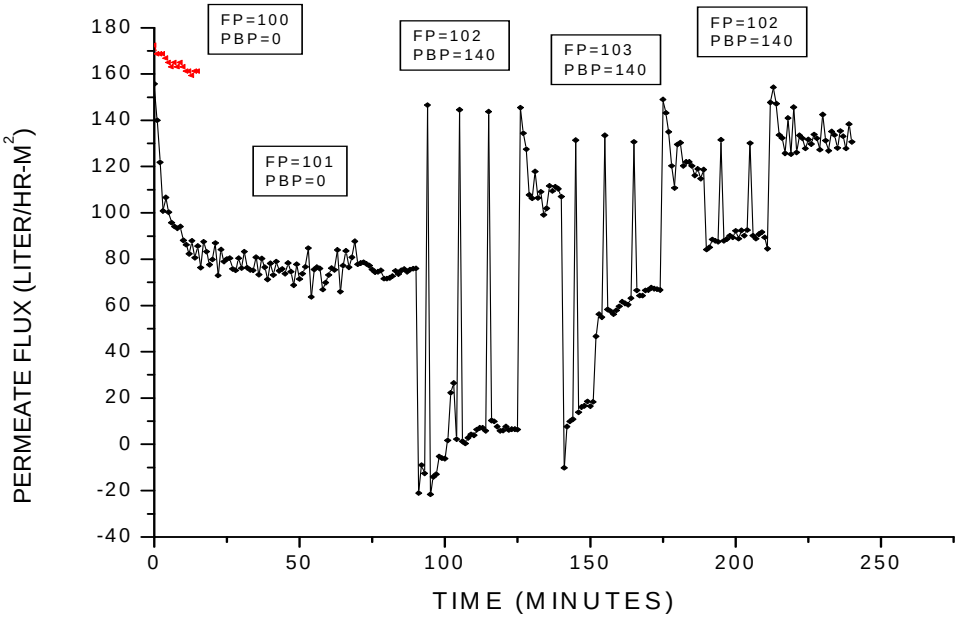


Figure 5.31 Flux against time for the PS membrane /yeast colloidal suspension system. (FP: feed pressure, PBP: peak of backpulse pressure).

CHAPTER 6

CONCLUSIONS

The main conclusions of this study are summarized as follows:

In all systems the first experiments involved cleaning the membranes by increasing the backpulse pressure from 90 to 140 and then to 180 kPa, using pure water as the source for the backpulsing pump, and soap solutions to investigate if the same or better cleaning could be obtained . The results of these experiments showed that in every experiment the permeate flux values, after each backpulsing cleaning cycle, increased with increasing backpulse pressure. The backpulsing with pure water at 180 kPa cleaned the membrane surface, but the apparatus could damage the flat sheet membranes. From these results it can be concluded that there was no need for soap solutions.

Experiments were performed with an alumina suspension during cross-flow MF, using a nylon membrane (0.2 µm pore size). Without backpulsing, the flux decreased due to the presence of alumina deposits on the membrane surface. With backpulsing with pure water, using three backpulsing cleaning cycles (all with peak amplitudes 140 kPa), the permeate flux values increased to 91, 93 and 95% of the initial pure water flux after the first, second and third backpulsing cleaning cycles, respectively. From the SEM results it can be concluded that backpulsing removed all of the fouling layer. This was supported by UTDR results. Backpulsing cleaning was however not very effective in removing fouling from inside the membranes. This was supported by measurements made using colloidal suspensions. This suggests that the mechanism of defouling was based on vibration of the membrane, which resulted in the removal of surface layer. Unblocking of pores was not observed, which indicated that reverse flow through pores was probably insignificant.

Experiments were also performed with a yeast suspension during cross-flow MF, using a nylon membrane 0.45 µm and cross-flow UF a 100,000 MWCO polysulfone membrane. During fouling operation, both internal and external fouling occurred. With backpulsing, with pure water alone (with peak amplitude 140 kPa), the permeate flux values for the 0.45 µm nylon membrane increased to 57, 56 and 56% of the initial pure water flux after the first, second and third backpulsing cleaning cycles respectively, while with the 100,000 MWCO

membrane they also increased to 91, 93 and 95%, respectively. From flux against time plots, UTDR measurements and SEM images results, it can be concluded that backpulsing removes nearly all of the fouling layers, but is not very effective in removing of internal foulant. This is supported by measurements made using colloidal suspensions. This again suggests that the mechanism of defouling was based on vibration of the membrane, which resulted in the removal of surface layer. Unblocking of pores was not observed, which indicated that reverse flow through pores was probably insignificant.

Experiments were performed during cross-flow UF of dextrin solutions with a 100,000 MWCO PS membrane. Results showed that, during fouling operation, a very thin foulant cake layer formed on the membrane surface after forward filtration for 60 minutes. This is supported by UTDR results and SEM images. After backpulsing, the permeate flux values increased to 97% of the initial pure water flux value after the three backpulsing cleaning cycles. From this it can be concluded that the backpulsing can effectively clean membranes fouling in UF which are used in this study.

When soap solutions were used in backpulsing, the following experimental results were obtained. In the case of MF of an alumina suspension through a nylon membrane; the final permeate flux values increased. In the case of MF of yeast through a nylon membrane and UF of yeast through a PS membrane, there was no improvement in the final flux values over using no soap. In the case of UF of dextrin through a PS membrane, there was also no improvement in the final flux values over using no soap.

The backpulsing with soap solutions never improved the final permeate flux values for filtration of all types of foulants used in this research. Moreover, it sometimes reduced the final flux values (in the case of washed yeast suspension with nylon 0.45 μm and PS membranes). The reason for this is that the soap could lyse the yeast cells to create debris which refouls the membrane during the periods when backpulsing is switched off in order to perform flux measurements. The results do not justify using backpulsing with soap solutions, because after use the soap solution has to be flushed out of the plant and disposed of which adds to the cost of cleaning when using backpulsing together with soap solutions.

In all cases, the flux values after each backpulsing cleaning cycle, UTDR measurements and SEM images, showed that the backpulsing always removes all (or almost all in the case of

washed yeast suspension with a nylon 0.45 µm membrane) of the caking fouling layer, but is not very effective in the removal of much of the internal foulant.

UTDR results showed that, most removal of the foulant layer is during the first backpulsing cycle. This is supported by permeate flux results.

Backpulsing through the membrane from the permeate side, with amplitude peak pressure of 140 kPa was found to be effective to clean the membranes. The permeate flux values after cleaning usually increased to the range of 60 to 98% of the initial pure water flux values.

UTDR was successfully applied to monitor membrane cleaning and evaluate the efficiency of the cleaning methods. The results showed that UTDR can measure the rate of cake layer formation on the surface of the membrane, using the amplitude and arrival time of differential signals as a function of operation time to provide information about the changes in the thickness and density of a fouling layer during forward filtration. From this it can be concluded that UTDR is very useful technique to understand the mechanism of fouling or the efficiency of cleaning procedures.