Removal of the Fouling layer on Membrane Filters used in Water Filtration by a Combination of Mechanical Impact and Purification Chemicals

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by

David McLachlan and Emad Shugman
University of Stellenbosch

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EXECUTIVE SUMMARY

Background and motivation

Membrane filtration has proved to be very successful for the treatment of wastewater, surface waters (fresh water and sea water) and groundwater (including brackish waters); it can be used to increase the quality of water and the quantity available. However, fouling is a major problem associated with membrane filtration; it results in a significant decrease in membrane flux. One of the major reasons for the decrease in efficiency of the filter materials (membranes) is the formation of a fouling or scaling layer on the surface of the membranes. However, before the fouling layer is formed, a nearly "instantaneous layer" is created by the foulant particles, or smaller particles associated with the foulant, partially blocking the pores. This takes place within much less than a minute of the foulant reaching the membrane surface. This manifests itself as a rapid drop in the flux, from the clean water flux value, in the first minute of fouling. This layer can prove more difficult to remove than the fouling layer itself which can be hundreds of microns thick. As it will reappear – even on a perfectly cleaned membrane – in the first minute of it being reused, one must, in certain circumstances, consider the flux after one minute to be the best flux value that can be obtained in practice.

The main aim of this research project was to study the cleaning of membranes by back pulsing them from the permeate space, both with clean (permeate) water and various soap solutions. A comparison was also made of the effectiveness of membrane cleaning obtained, using only water and various soap solutions. It should be noted that final flux values obtained using soap solutions must be significantly higher than those obtained using clean permeate water to justify the more complicated procedures that are entailed in using soap solutions. Also the additional cost of the soaps and the problems and expense that will be encountered in disposing of these soap solutions must be considered.

There are several reasons why this research program was considered to be important and should be carried out in South Africa and at Stellenbosch in particular:

South Africa is a water-scarce country with a great need for purifying surface water and recycling industrial effluents.

The need for membrane filtration systems exceeds the current base of filtration plants and a more effective method of cleaning of membranes will lead to an increase in the running time of existing plants.

The improvement of the membranes themselves requires a better basic understanding of the fouling process; which can be aided by this study of the cleaning of

membranes. It can be noted that membrane development for water treatment is one of the fortes of South African researchers.

A further excellent reason for motivating this study is that the Ultrasonic Time Domain Reflection (UTDR) technique, which can be used to study the formation of the fouling layer, is only available at a limited number of laboratories worldwide, one of which is at Stellenbosch.

South African water treatment companies should benefit from the results of this research.

Objectives and aims

Aim 1

The modification of the existing apparatus (as used in WRC Project K5/1441) by:

1. The addition of a back pulsing pump and the necessary plumbing modifications.

2. The addition of new pressure sensors and upgrading the LABview data acquisition system.

3. Making provision for the introduction of the soap solutions into the plant.

This was achieved and a plan of the new plant is included in the report.

Aim 2

Initial and final tests of the effectiveness of the cleaning using different membranes, effluents (foulants) and back pulsing sequences.

1. The membranes used were an Alfa Laval GRO Polysulphone 100 000 molecular weight cut-off (MWCO) membrane and two PAL Biodyne nylon membranes with specified pore sizes of 0.2 and 0.45 μm.

2. The foulants used were:

    a. Alumina powder, which is chemically inert and had a particle size of 1 μm (with a small size distribution).

    b. Commercial yeast powder, which consisted of 10 μm cells that also had adhesive properties and which could “plug” the 0.45 μm nylon membrane pores. A difficulty with this live yeast was that it continually produced proteins and other colloidal materials.

c. Dextrin, which consisted of a range of small molecules, some of which penetrated the membrane, and has strong adhesive properties.

3. The soap solutions used were:

   a. A SMBS cleaning solution: 1 g sodium lauryl sulphate, 1 g calcium hypochloride and 1 g ethylene diaminetetraacetic acid in 1 ℓ of reverse osmosis (RO) water.

   b. A Polystep F9 soap solution (nonylphenol ethoxylate): 1 g in 1 ℓ of RO water.

4. Various sequences of cleaning back pulses were used, which are detailed in the report.

5. Ultrasonic time domain reflection (UTDR) spectroscopy was used to measure the thickness of the foiling layers.

Over 50 experiments were carried out to test the effectiveness of back pulsing, with and without soap solutions, in cleaning the above membrane with the above model foulants. Most of these are detailed in the main report as flux versus time curves, together with the results of the evolving thicknesses of the foulant layer using UTDR.

Aim 3

Analysis of the membranes, before and after use, to improve our understanding of the cleaning process and the membranes:

1. The most useful of the techniques, mentioned in the proposal, was an examination of new, fouled and cleaned membranes using scanning electron microscopy (SEM).

2 Atomic Force Microscopy, which proved to be far too sensitive to obtain useful results from the fouled and cleaned membranes. No results are attached.

3. Infrared analysis, which did not give any meaningful results as the beam was focused by the manufacturer to probe several hundred microns under the surface. This meant that even for a fully fouled membrane, the spectra obtained were primarily that of the membrane material.

4. Ultrasonic time domain reflection proved useful not only in measuring the thickness of the fouling layer, but also useful information as to the final state of

the membrane was gained from the difference in pulse shapes between the original UDTR signal and the final one.

About 20 representative SEM images where taken of new, fouled and cleaned membranes. These proved to be extremely useful in interpreting the flux-time curves and the results of this interpretation are briefly given below and in more detail in the main report.

Results and conclusions

A description of the experimental apparatus and the procedures is given in the main report, where the results of all the experiments are summarized in two tables and over twenty figures. A number of examples of flux-time curves and, in some cases, the relevant UTDR results are given, as are a number of SEM images.

The first set of experiments involved cleaning the membrane with successive 90, 140 and 160 kPa pulses of 30 minutes each, using both permeate water and soap solutions. At 140 and 180 kPa these experiments showed that the back pulsing increased the flux rate. The increase was largest after the 180 kPa pulse and there was very little difference between back pulsing with and without soap solutions. At this point the conclusion could be drawn that back pulsing at 180 kPa could very satisfactorily clean the three types of membranes used of the three foulants listed above. However, as continuous back pulsing at 180 kPa could result in long term damage in spiral wrap membranes, an investigation as to whether the same or better cleaning could be obtained at lower back pulse pressures by using soap solutions (see Table 2 in the main report).

A sequence of three 30-minute 140 kPa back pulses consisting of a first pulse using permeate water, a second with a soap solution (to see if this increased the flux above that obtained by the first pulse) and finally a third back pulse using permeate water to wash away any residual soap (and debris) that may still have been on or in the membrane, was used. Naturally these had to be compared with three successive RO water pulses. These experiments were carried out for four membrane and foulant combinations, which were PAL 0.2 μm/alumina, PAL 0.45 μm/yeast, GRO/yeast and GRO/dextrin. The results are given in Table 2 in the main report. Unfortunately, as the results were not as reproducible as was expected, many of the experiments had to be repeated. In addition, the SMS solution, as given above, was tried at one half and double concentrations in some instances. All this resulted in a large number of experiments being performed.

The PAL 0.2 μm/alumina system was probably cleaned best by the back pulse method. Surprisingly, due to the inertness of the alumina, the small improvement using the soap solutions was probably the best for any of the systems.

The results for the PAL 0.45 μm/yeast systems were erratic but no significant difference in the flux values, with and without soap solutions, could be detected and certainly not enough to justify the use of soap solutions.

The GRO/yeast system results showed that the use of soap solutions could actually make the final flux value worse. Note that an ultra filtration membrane would not normally be used to filter a large cell like yeast but was introduced to compare with the PAL 0.45 μm/yeast and GRO/dextrin systems.

The GRO/dextrin results were again erratic, but the results using soap solutions were not significantly different, from the permeate water only sequence, to justify using soaps in this system. It should be noted that the fouling layer could not be clearly detected by US measurements but its formation was quite clear from the flux-time results. The fouling layer was also not very apparent on the SEM image of a fully fouled sample.

In all cases the UDTR measurements showed the fouling layer had been wholly or partially removed by the first back pulse of 30 minutes. This meant that the subsequent pulses were only further cleaning either the foulant in the pores or a very thin remnant layer on the surface, or both. However, as the flux in several experiments dropped between the first (BP1) and the BP2 and BP3 pulses, it has to be concluded that in some cases the subsequent back pulses had a negative effect on the surface of the membrane or its supporting pore structure. In all cases the shape of the UDTR pulse reflected from the cleaned membrane was different from that of the original pulse, showing that some foulant material was imbedded in the pores, and/or that there were some particles still adhered to the surface and/or that the membrane had been distorted. The first two observations were usually reinforced by the SEM images taken from the cleaned surfaces. Note that, the UDTR reflections from the cleaned dextrin surface (which often showed a higher cleaned flux value than the original new membrane) were also not the same as the original or new membrane reflection. The SEM image for dextrin showed no adhered layer or material in the pores, which is to be expected as dextrin molecules could not be resolved by the SEM. Therefore, the possibility remains that the membrane has been distorted by the back pulse.

The SEM images of the new membrane surfaces were consistent with what could be expected, the pores in the GRO surfaces not being visible at a magnification of ×4000. The SEM images of the fouled surfaces were consistent with the size of the foulant but somewhat rougher than was expected. An exception was the dextrin surface which was very similar to that of the new surface, but as the fouling layer was very thin and the molecules cannot be resolved by the SEM, this is not surprising.

The SEM images taken after 15 seconds gave an indication of the original, or "instantaneous", fouling process. In the case of yeast and a PAL 0.45 μm membrane it can clearly be seen that the yeast cells were plugging the pores and in the case of the GRO membrane they are clustered around the (invisible in the SEM) most effective pores. For alumina all that can be seen is that the particles are clustered around some of the "0.45 μm" pores, which must those at the entrance of the most effective percolation paths.

The SEM images of the final cleaned surface could usually be correlated with the different final flux values, in that there were more particles in the pores and/or on the cleaned surfaces of the ones that showed a lower final flux value.

The summarized conclusions are:

Back pulsing through the membranes, with peak pressures of 140 kPa and more, very effectively cleaned the membranes, with new clean water values usually being in the range of 70 to 100% of the original RO value. The main action of the back pulse was to remove all, or a significant fraction, of the fouling layer but it could also have washed some colloidal matter out of the pores. In most cases the final flux was higher than the flux after less than one minute of fouling, which was primarily due to the instantaneous layer, showing that the back pulsing had partially removed the instantaneous layer. A remnant of the surface layer could be seen in some of the SEM images. It was not clear to what extent the instantaneous flux and the final cleaned value of the flux were influenced by colloidal particles trapped in the pores, but experiments with colloidal foulants showed them to be very effective foulants.

Using soap solutions during the back pulsing never significantly increased (it sometimes decreased) the final cleaned value significantly. Certainly the improvement was never enough to justify the additional trouble and expense of using soap solutions.

Future developments

This project was a test case for further cleaning and continuous back pulsing studies, using spiral wrap membrane elements. The study showed the directions that future work on back pulse cleaning of capillary membranes should take. As well as looking for improvements in the cleaning procedures for filtration elements, similar studies could involve both continuous and/or intermittent back pulsing to keep the membrane at a high flux level for long periods.

As the proposal emphasized the use of soap solutions to improve the cleaning obtained using back pulsing, the experiments focused on this aspect. Due to the

nature of the proposal, time constraints and the rather irreproducible data it was not possible to extend this study to include other possibilities.

Future work could include passing the cleaning solution over the fouled layer while back pulsing with permeate water from below. From the experiments reported on here, it is almost certain that the fouling layer would be near-instantly lifted and disintegrated by the through-membrane back pulse. The soap solution would then pass directly over the membrane and could possibly lift some of the foulant and debris left by the back pulsing. However, it is hard to visualize why the soap solution flowing over the membrane should be more effective than the same solution passing through the membrane.

ACKNOWLEDGEMENTS

We would like to thank the WRC for the financial support of this project.

The authors of this report would also like to acknowledge the groundwork undertaken by Dr Frank Reinecke in initiating this project, prior to his untimely demise.

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1. INTRODUCTION

Membrane filtration of aqueous effluents is a widely used technique in water purification in the food industry, to clean industrial effluents prior to discharge or re-use and to obtain potable water from various sources. The fouling of membrane filters during the filtration process of all aqueous effluents is a well known phenomenon. Fouling of the membranes causes a decrease in permeate flux and eventually the filtration process becomes so inefficient that a cleaning procedure must be initiated. The application of pulses in the infrasound regime as all or part of the cleaning procedure is studied, to see if it improves the efficiency and lowers the cost of cleaning procedures.

As previously stated, fouling of the membranes causes a rapid, disadvantageous decrease in permeate flux [1,2]. It is mainly the sub-micron proteins and polyphenols, i.e. smaller than the membrane pores, which are considered to be the cause of this initial membrane fouling [2]. After this the flux decreases due to the formation of a caking layer. Then, in order to restore permeate flux, the filtration process has to be interrupted to clean or to replace the membrane filters. For economic reasons membrane cleaning which does not seriously interrupt the filtration process or use hard to dispose of chemicals is preferable.

There are a number of such membrane cleaning techniques available, including back flushing, cross flushing, and back pulsing (or back shocking) [3-9]. Although these techniques, applied in a non-continuous way, could be successfully used to clean membranes and restore the flux, they seem to be inefficient in the removal of adhesive foulants [10]. Previously [2, 10], and in this paper, rapid continuous back pulsing from the permeate space into the feed space was found to be very effective. In order to avoid back-side contamination of the membrane, a clean feed (the permeate itself or reverse osmosis (RO) water) had to be used for the back pulsing. Czekaj et al. [2,10] used infrasonic back pulsing at frequencies of 5-10 Hz for such a purpose. They filtered 0.66 g/ℓ washed yeast and 0.5 g/ℓ talc suspensions through flat-sheet filters of polyvinylidene difluoride (PVDF) membranes. Compared to the "steady state" fluxes achieved with standard cross-flow filtration, the permeate fluxes, with continuous back pulsing, were four times higher for the talc suspension and three times higher for the yeast suspension [10]. As such infrasonic back pulsing from the permeate space shows considerable potential for flux enhancement, its potential to aid in the cleaning process is investigated in this paper.

Ultrasonic time-domain reflectometry (UTDR), an *in-situ* non-invasive, real-time measurement, provided information about the growth of the fouling layer on a membrane [11-15]. As a direct correlation between the growth of the fouling layer and the decrease in the permeate flux existed, the UTDR technique was used to help monitor the efficiency of the cleaning process.

The primary study in this paper consisted of measuring the flux as a function of time, before and after fouling as well as before and after various infrasonic back pulse sequences had been applied to the fouled membrane. The back pulses were applied to the membrane from the permeate space and could consist of either permeate water or a soap solution. As part of this investigation the fouling of the membrane was also monitored in situ by ultrasonic time domain reflectometry (UTDR). Scanning Electron Microscope images of some of the membranes were made on new membranes and fouled membranes, as well as on membranes recovered, at various stages of the cleaning process.

As there is no appropriate theory for this process, the experimental apparatus is described in the next section. The procedures used will be given in Section 3 and the results and discussion will be presented in Section 4. The conclusions are made in Section 5 and suggestions for future work in Section 6.

2. THE EXPERIMENTAL SYSTEMS

2.1 Membranes and Foulants

The membranes, foulants and soaps used are as follows:

Membranes:

Alpha Laval GRO 100 000 MWCO membranes	G in the Tables
A PAL Biodyne 0.45 μm nylon membranes	P1 in the Tables
PAL Biodyne 0.20 μm nylon membranes	P2 in the Tables

Foulants:

Dextrin (0.5 g/ℓ)	D in Table 1
Cleaned yeast* (1g/ℓ)	Y in Table 1
Alumina powder** (1g/ℓ), 1 μm diameter with small size distribution	A in Table 1

*The yeast was rinsed with RO water and centrifuged for 8 mins and the colloids in suspension were discarded. The process was repeated a further two times.

**The alumina was shaken with water, which then had a cloudy appearance. After the alumina had settled the colloidal matter was poured off. The process was repeated once.

2.2 Cleaning Solutions/Soaps

SMBS cleaning solution; 1 g sodium lauryl sulphate, 1 g hypochloride and 1 g ethylene diaminetetracetic acid in 1 ℓ of RO water.	S in the Tables (S/2 indicates a dilution of 2 and 2S a more concentrated solution by a factor of two.)
Polystep F9 soap solution (nonylphenol ethoxylate); 1 g in 1 ℓ of RO water.	F in the Tables

A rectangular, flat-bed micro filtration module (Figure 1) of 100 mm length, 32 mm width and 2 × 19 mm height, was used in all the flat-cell experiments. The module was made of polymethyl methacrylate (Perspex) and consisted of two plates (each 19 mm thick, 200 mm long and 94 mm wide). The feed space cavity in the top plate (2 mm deep, 88 mm long and 30 mm wide) has three 2 mm pipes at each end which, through individual manifolds, were connected to the input and output feed lines. The membrane rested on a sintered brass plate, set in the lower Perspex plate, and below this was another cavity, 13 mm deep, 88 mm long and 30 mm wide, to collect the permeate. The effective membrane area was 0.0032 m^2 (100 mm × 32 mm), which is the area of the brass sintered plate.

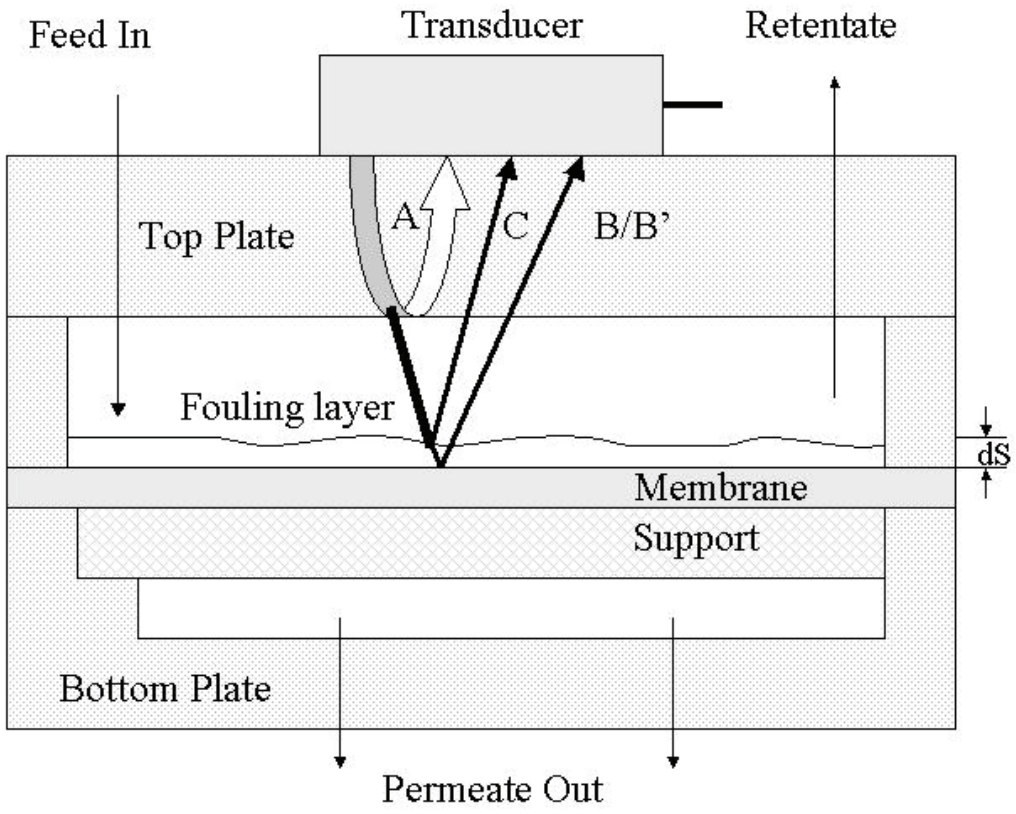

Figure 1. Schematic representation showing a cross-section of the cell and the principle of UTDR measurement in a flat-bed membrane module [15]. (All the monitored incident and reflected acoustic waves (A, C, B/B') were perpendicular to the transducer and the membrane surface).

The ultrasonic measurement system (Figure 2) consisted of a 7.5 MHz ultrasonic transducer (Panametrics V120), a pulser-receiver (Panametrics 5058PR), and a digital oscilloscope (HP Model 54602B, and later a Tektronix Model TDS 2024). The pulser-receiver setup had the following specifications: bandwidth 10 MHz, repetition rate 200 Hz, damping 50 ohms, pulse height 200 V, attenuator 25 dB, gain 60 dB, high pass filter – out, low pass filter – out, mode – normal. The oscilloscope had a 150 MHz bandwidth, 1 mV/div vertical sensitivity, and an 8-bit digitizer. The sampling frequency was 250 MHz. The transducer was externally

mounted with a bracket into a close-fitting cavity on the top of the cell, thus maintaining exactly the same distance between transducer and cell in each experiment. Commercially available 'high viscosity ultrasound gel' was used to couple the transducer to the surface. The pulser-receiver generated the required voltage signal to trigger the transducer, which sent out an ultrasonic pulse. The oscilloscope captured and displayed the returning signal amplitude (volts) as a function of arrival time (seconds). Each set of ultrasonic signals generated consisted of 500 data points. A computer (PC) was connected to the oscilloscope to store the data at the required intervals.

Two peristaltic pumps (Watson Marlow 323 and Watson Marlow 313), operating at constant flow, were used to supply the feed to the cell. Feed-pressure changes were displayed digitally and recorded by the LabView programme. The pumps were connected to a three-way valve for rapidly changing the feed from the clean water, used for conditioning of the membrane, to the foulant in the input tank of the pump.

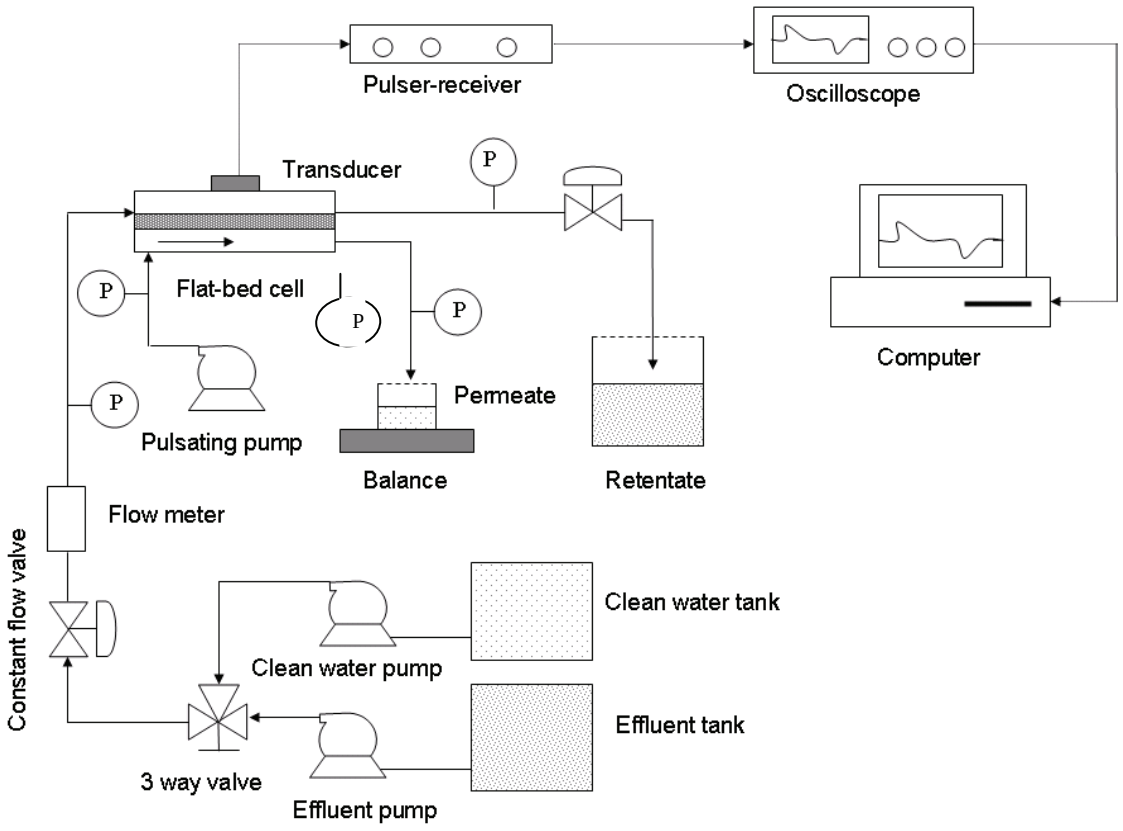

Figure 2. Schematic representation of experimental set-up of the microfiltration separation and ultrasonic measurement systems used.

For the infrasound/pulsing a diaphragm pump was connected to the permeate side of the cell. The frequency of the back pulsing was 5 to 6 Hz. Figure 3 shows typical pressure-time traces

of a pressure pulse at the beginning (green) and end (red) of the first 30 minutes of back pulsing at 140 kPa.

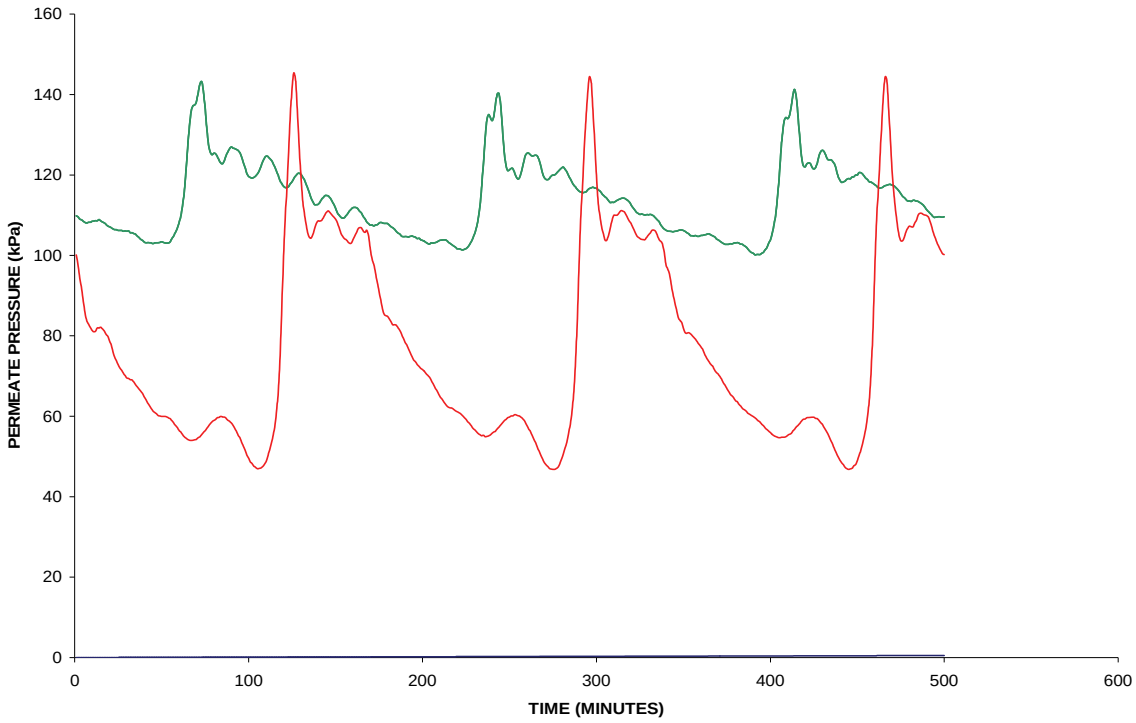

Figure 3. Pressure amplitude against time plots, in permeate space, at the beginning (green) and end (red) of the first 30 minutes of back pulsing.

The permeate flow was fed into a large beaker on top of an electronic balance and the mass was measured using an electronic balance. The mass change in a selected period was divided by the period to ascertain the flow rate. The period was selected by the operator and could be as short as 5 seconds during the initial fouling, but was always 30 seconds after the first minute or two. During back pulsing the liquid for the back pulsing pump was taken from the beaker on the balance so that only the nett increase in mass, which was due to the permeate flux, was recorded.

3. EXPERIMENTAL PROCEDURES

In order to follow the discussions below Figures 4 to 10 should be consulted.

Step 1. In all experiments the membrane clean water value (100%) was measured first using RO water with a positive trans membrane pressure (TMP) of 100±1 kPa and a cross flow rate of 45±3 ℓ/hr for a period of 10 minutes. Using the Labview system, the feed pump was then changed so that a fouling solution was introduced in the feed space at a pressure of 100±1 kPa, a flow rate of 45±3 *l*/hr and a cross flow velocity of 0.25 m/sec. The switchover time became the zero time in all experiments (refer to Figures 4-10). In all cases a rapid

decrease in the flux was observed over the first 5-15 seconds, due to the foulant (or colloidal) particles blocking the pores. After this, the flux dropped at a far slower rate, as the foulant/caking layer formed. This behaviour can be seen as an elbow, close to the origin, in the flux-time plots given in Figures 4-10. After fouling for 60 minutes the intake of the pump was changed to RO water, again at 100 kPa, in order to wash any loose matter off the fouling layer. In all cases only small or no changes in the flux were observed.

Step 2. The membrane was back pulsed for 30 minutes, from the permeate space, using either RO water or a soap solution as the source for the back pulsing pump. The back pulsing frequency was 6.7 INSERT UNIT and the peak pressure was chosen or varied for a particular experiment. Therefore negative TMPs occurred during the pulse cycle and negative flux values were possible during back pulsing. Note that flux percentages are obtained from the flux at the point in question divided by the clean (RO) value observed for that particular experiment.

During all back pulse periods the back pulsing was stopped for three 1 minute intervals to check the then current “clean” water flux. The back pulsing was then switched off and a new clean water flux value (BP1) was measured over the next 15 minutes using RO water. (The input tank, containing either RO water or the fouling solution, for the pulsating pump was changed where necessary). All results up to this point depended only on the type of membrane being used. The statistical error is rather large, but this could be due to the small active area of membrane, cut from a large sheet.

Step 3. The system was back pulsed for a further 30 minutes, usually at 140 kPa using either RO water or a soap solution. The back pulse pump was switched off and a new clean water flux value (BP2) was then measured. Experiments in which the first pulse was made with a soap solution terminated at this point.

Step 4. If necessary, the membrane was again back pulsed using RO water for 30 minutes, after which a new clean water value (BP3), again using RO water, was obtained in the last 15 minutes of the run.

The membranes at the end of an experiment were stored in a sodium metasulphate solution, before some of them were examined using SEM.

For SEM work a small section (approx. 5 × 5 mm), was cut from the centre of the wet membrane and allowed to dry. It was then mounted on a SEM stub. Up to 8 stubs could be measured at a time. Images were taken at magnifications of ×500, ×1500 and ×4000. The lower magnifications, which are not shown, were used to check that the areas examined at ×1500 and ×4000 magnifications were typical of the section of the membrane.

4. EXPERIMENTAL RESULTS AND DISCUSSION

A few two- and three-pulse sequence experiments were performed in which all the peak pulse pressures were below 100 kPa. In all cases the cleaning of the membranes was negligible, or far below what would be considered worthwhile in practice, and so the results are not presented here.

4.1 Experiments using a 90 kPa, 140 kPa, 180 kPa sequence (Figures 4 to 6 and Table 1)

Step 1 was done at a peak back pulse pressure of 90 kPa, Step 2 at 140 kPa and Step 3 (when applicable) at 180 kPa. In these early stages, some of the experiments were terminated at Step 2. It was only after the cell had been modified with a spacer cloth over the membrane, to limit outward bulging, that pressures of 180 kPa (Step 3) were used. Unlike the experiments presented in section 4.2, the RO or the soap solution indicated on the figure was used in all the back pulses and in arriving at the BP1, BP2 and BP3 flux flow results for these experiments.

Representative results are presented in Figures 4, 5 and 6 and summarized in Table 1.

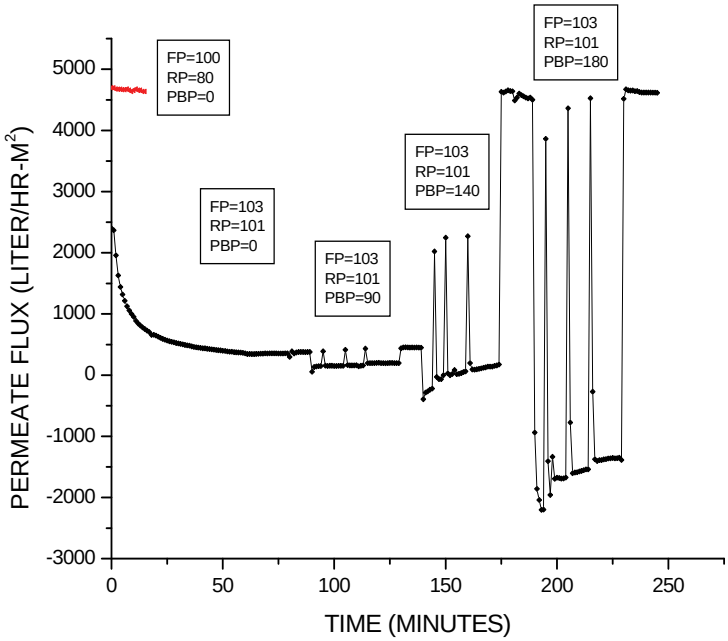

Figure 4. Plot of the flux against the time for the PAL 0.2 µm/alumina system.

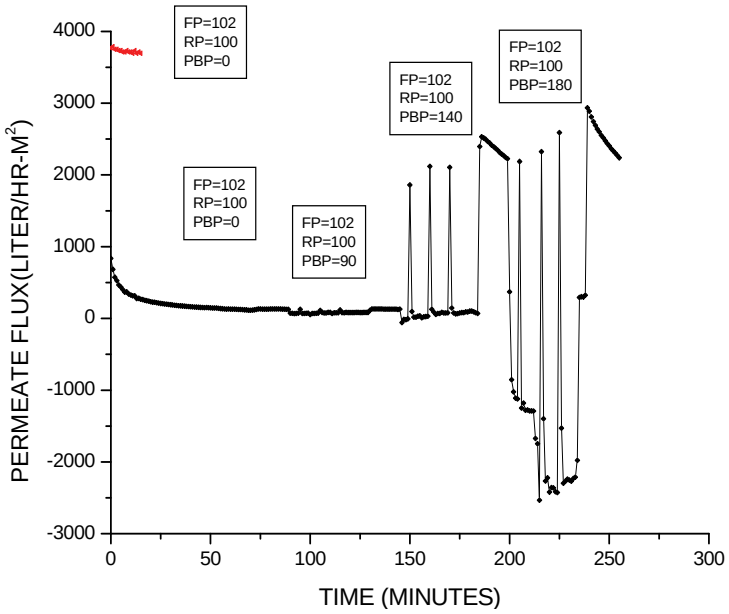

Figure 5. Plot of the flux against the time for the PAL 0.45 µm/yeast system.

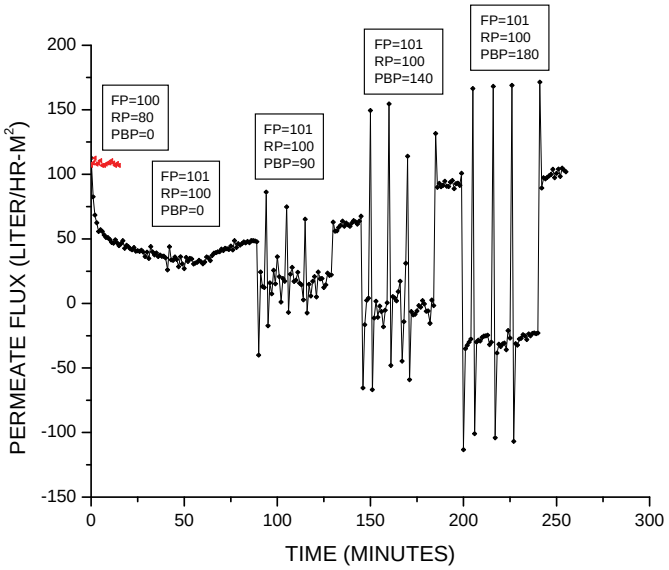

Figure 6. Plot of the flux against the time for the GRO/yeast system.

From these preliminary results it was concluded that, as no clear differences between the RO water and the soap solutions were discernable, another sequence of back pulses had to be used to ascertain if there was any difference. It was also noticed that the flux after BP3 was always higher than after BP2.

Table 1. Flux results for experiments described in Section 4.1.

Membrane /Foulant*	RO (after 5 min using RO water)	5 MIN (after 5 min)	60 MIN (after 60 min)	90 MIN (after cleaning)	PBP 90 (after 90 kPa back pulse)	PBP 140 (after 140 kPa pulse)	PBP 180 (after 180 kPa pulse)
P2/A-R	240	70	18	20	24	239	239
P2/Y-R	97	80	3.11	3.1	14.2	18.3	21.1
P2/Y-S	140	90	4.6	1.42	1.18	1.26	4.77
P1/Y-F	50	30	3.63	5	4.4	18.6	12.3
P1/Y-R	200	23	6.9	6.8	7	126	130
G/Y-R	15.4	10	4.9	4.3	4.3	10	
G/Y-R	6.1	5	1.7	2.55	3.2	5.1	5.5
G/D-R	10.15	8.9	2.5	3	3.8	4.7	
G/D-R	11.9	9.2	2.5	2.8	6	5.4	
G/D-R	8.4	7	2.7	3.75	5.4	5.4	6.5
G/D-S	8.75	7.3	2.6	2	2.1	5.2	
G/D-S	9.4	8.2	2.5	1.84	2.3	4.5	
G/D-F	9.2	8.5	2.6	1.2	2.6	7.75	
G/D-F	10.5	8.1	2.9	1.5	4.2	4.2	9

*G GRO 100 000 MWCO; P1 PAL 0.45 μm nylon; P2 PAL 0.20 μm nylon
*D Dextrin; Y Cleaned yeast; A Alumina powder
*S SMBS cleaning solution; F Polystep F9 soap; R RO water

4.2 Experiments using a three 140 kPa pulse sequence (Figures 7 to 10 and Table 2)

In these experiments, Steps 1, 2 and 3 were all carried out at 140 kPa. After fouling and washing the membrane, the back pulsing in Step 1 was carried out using RO water, Step 2 with RO water or a soap solution and Step 3 with RO water again. The flow rates after BP1, BP2 and BP3 were always measured using RO water.

A 30-minute 180 kPa back pulse always improved the flux. The rationale for this procedure was to see if the second 30-minute soap solution back pulsing could improve the flux over that already obtained using the first 140 kPa back pulse, and also after that obtained with a second RO water back pulse. If this were not the case, it would not be justified to use a soap solution, with all the attendant problems.

Representative flux-time plots are given in Figures 7 to 10 and the results summarized in Table 2. Note that the results up to and including the first back pulse are dealt with first, as they should depend only on the membrane being studied.

In all cases, and more markedly for the two nylon membranes, a rapid decrease in the flux was observed over the first 5-15 seconds, due to the foulant particles blocking the pores. The flux then dropped at a far slower rate, as the foulant/caking layer formed. This behavior was

seen as an elbow, close to the origin, in flux-time Figures 7-10. After fouling for 60 minutes, RO water was introduced into the feed space at 100 kPa for 30 minutes to wash any loose matter off the fouling layer. In all cases only small changes in the flux were observed.

The membrane was then back pulsed from the permeate space for 30 minutes, using RO water as the source for the back pulsing pump. As the peak pressure was 140 kPa, negative flux values are possible during back pulsing, as can be seen in Figures 7-10. After this a new clean water flux value (BP1) was measured over the next 15 minutes. From Table 2 it can be seen that for the PAL 0.45 μm membrane the BP1 values lay between 43 and 105% and values for the GRO membrane lay between 75 and 90%. The reasons for this are not understood, nor are the reasons why the BP1 values for the PAL 0.2 μm membrane lie between 43 and 122%. The system was then back pulsed for a further 30 minutes using either RO water or a soap solution before the back pulse pump was switched off and a new clean water flux value (BP2) was measured (Step 3).

Finally the membrane was again back pulsed using RO water for 30 minutes, after which a new clean water value (BP3) was obtained in the last 15 minutes of the run (Step 4).

As the BP2 and BP3 results depend on both the foulant and membrane, they will be discussed separately below.

Again in both cases the statistical error was rather large but this could have been due to the small area of membrane cut from a large sheet.

The results in Table 2 are summarized graphically in Figures 11-14, where the fluxes at various times are plotted vertically for each membrane as numbered in Table 2. It was hoped that these plots would help clarify the erratic behavior of the flux values.

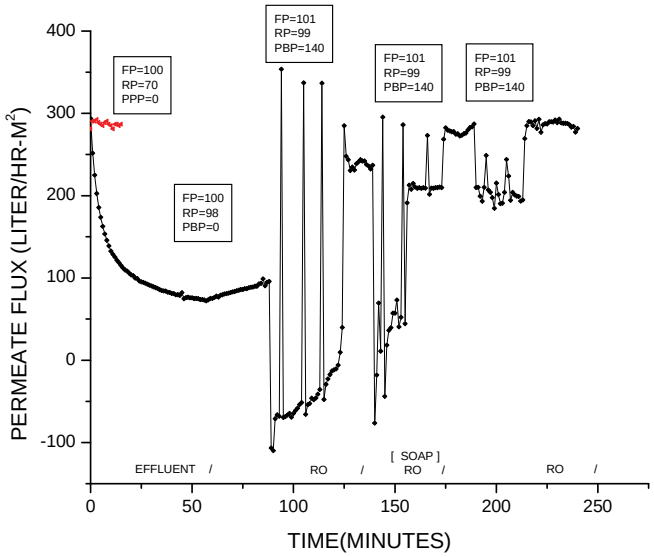

Figure 7. Plot of the flux against the time for the PAL 0.2 µm/alumina system

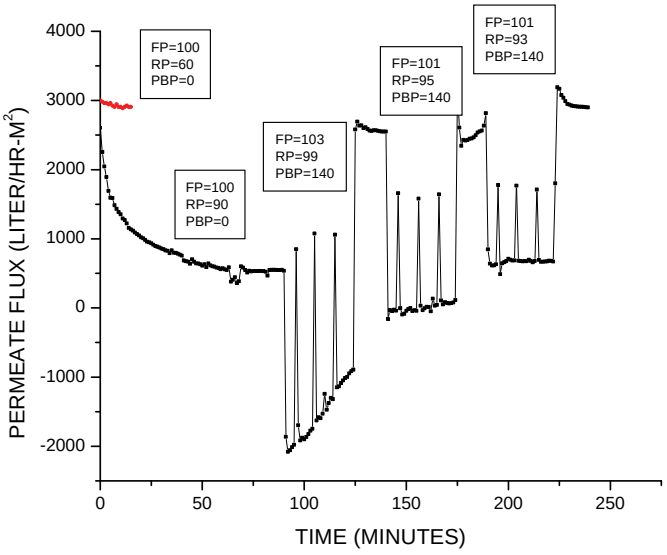

Figure 8. Plot of the flux against the time for the PAL 0.45 µm/yeast system

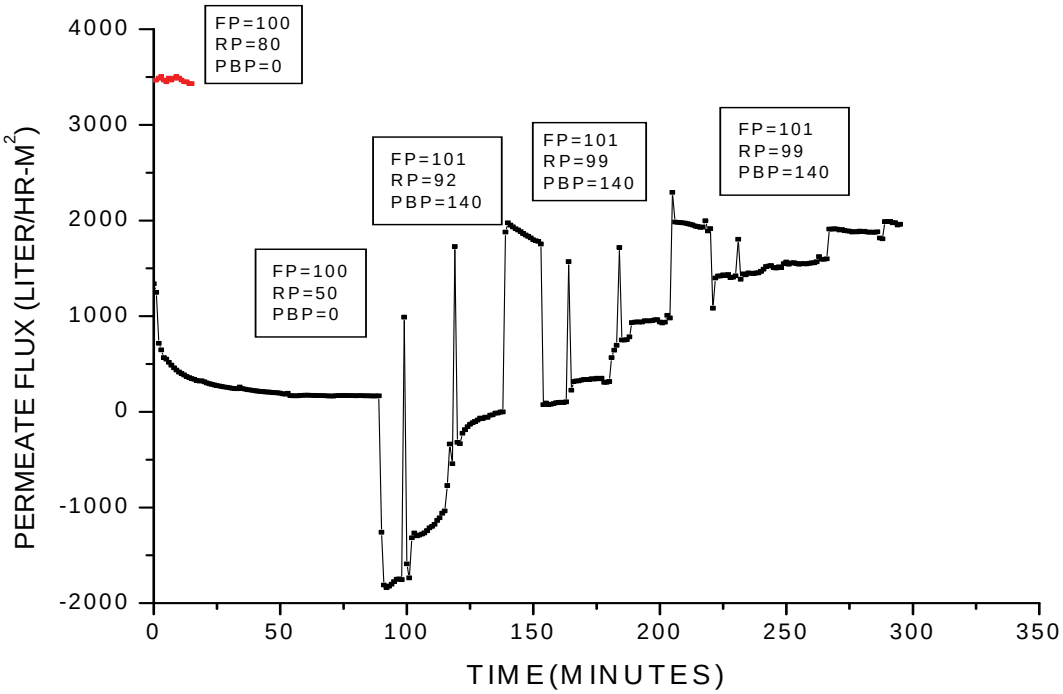

Figure 9. Plot of the flux against the time for the GRO/yeast system

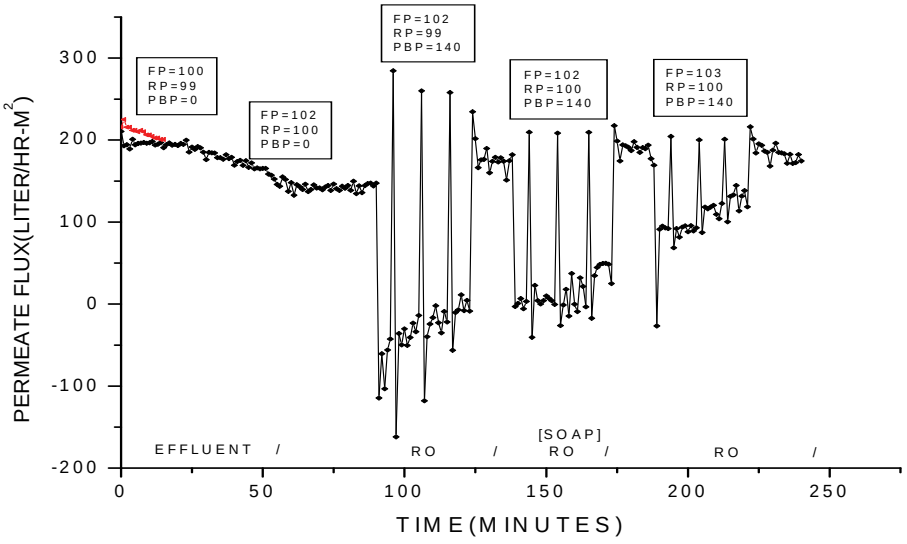

Figure 10. Plot of the flux against the time for the GRO/dextrin system

Table 2. Flux results for experiments described in Section 4.2.

Membrane / Foulant[1]	RO	5 MIN	60 MIN	90 MIN	BP1	BP2	BP3
P2/A-R	155	63	20	20.5	152	150	105
P2/A-R[2]	160	85	29	29	136	136	156
P2/A-R	156	62	12	22	160	167	165
P2/A-S	140	80	21	17.5	172	170	172
P2/A-S[2]	160	66	24	24	70	190	165
P2/A-F	142	75	25	25	168	158	164
P1/Y-R[2]	150	36	9.2	8.8	111	54	33
P1/Y-R[2]	180	30	9.2	8.9	104	105	106
P1/Y-R	185	23	6	5.1	130	99	70
P1/Y-S0.5	187	24	5.5	4.7	196	140	40
P1/Y-S1	184	30	6.7	6.2	107	118	97
P1/Y-S1[2]	170	26	8.2	7.2	73	112	101
P1/Y-S 2	190	22	6	5.8	197	80	30
P1/Y-F	155	30	8.3	9	168	112	56
G/Y-R[2]	12.3	10.3	3.4	3.4	10	10.7	11
G/Y-R	11.5	10.7	3.8	4.2	9	10.2	10.5
G/Y-S0.5	11	9	3.2	3.5	7	7.4	7.4
G/Y-S1[2]	9	9	3.3	3.2	8.6	8	7.6
G/Y-S 2	11.8	10.7	3.5	3	6.8	6.4	5.8
G/Y-F	11.6	9.8	3.3	3	9.5	6.3	6.8
G/D-R[2]	12.9	9	4.5	5.4	11.8	11.8	9
G/D-R	14	9.2	3.8	4.2	12.7	13.6	13.7
G/D-R	12.6	9.1	3.6	4.5	10.8	11.4	10
G/D-R[2]	15	9.2	4	5.1	13.5	14.5	15.4
G/D-S	11.9	9.4	4.9	5.1	10	10.6	12.5
G/D-S	11.7	8.5	4.6	5.4	9.7	11.6	12.4
G/D-F	10.5	8.1	4.3	5.3	9.6	11.5	11.8
G/D-F	11.4	8.7	4.6	5.8	8.6	7	8.8
P1/YM	170	24	4	4.7	55	39	23
P2/AM	170	80	20	20	41	25	20
G/YM	9	5.3	3.9	4	6	6.4	6.8
P1/Y-Q	185	22	6	5.3	162	113	106
D/Y-R	16	15	5.6	4.8	11.5	10.8	11.2
P2/Y-R	162	23	5.6	5.5	60	31	21
G/D-S	12.3	8.9	3.3	3.9	10.6	11.5	
P1/Y-S	187	22	6.5	5.3	120	76	
G/Y-S	12.2	7	3.1	3.7	10.7	11.4	
P2/A-S	157	69	24.8	26.5	160	155	

[1]G GRO 100 000 MWCO; P1 PAL 0.45 μm nylon; P2 PAL 0.20 μm nylon

D Dextrin; Y Cleaned yeast; A Alumina powder

S SMBS cleaning solution; F Polystep F9 soap; R RO water

[2] A SEM image has been taken of this experiment.

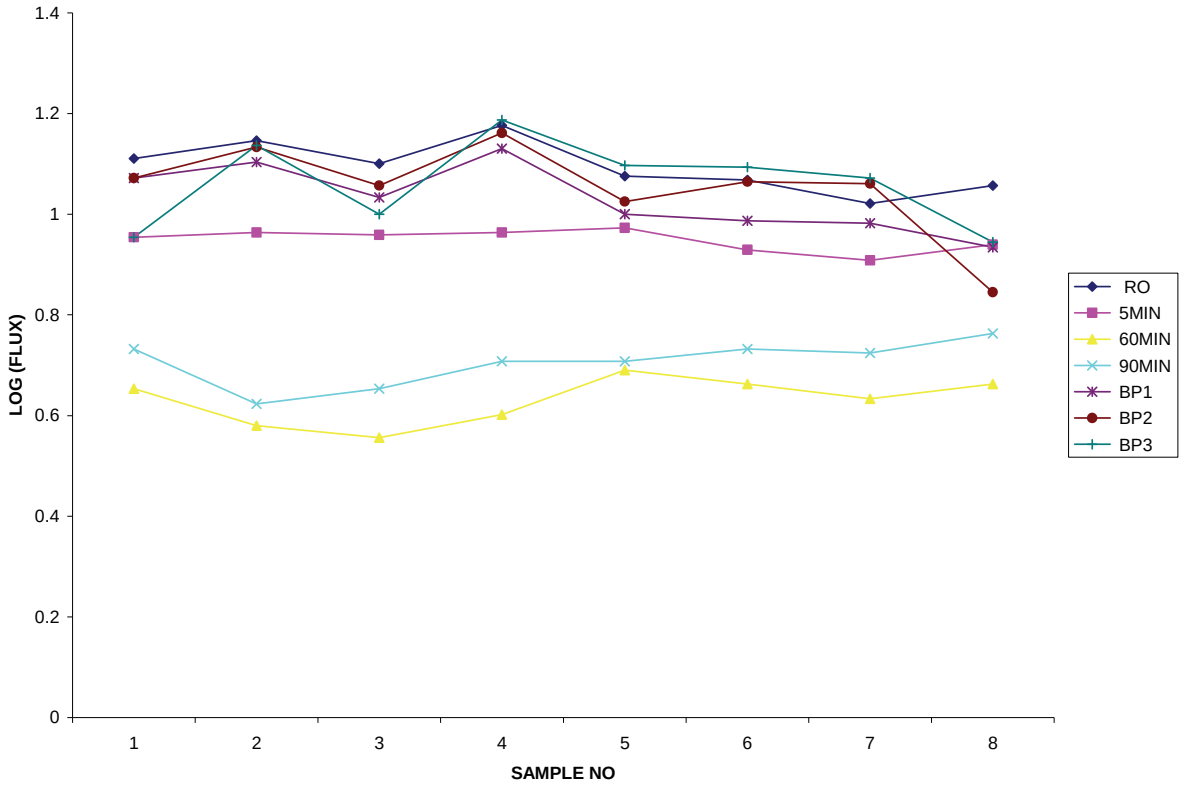

Figure 11. Plot for the PAL 0.2 µm/ alumina systems of the flux at the intervals specified in Table 1.

A) PAL nylon 0.2 µm membrane with alumina (Figures 7 and 11)

Table 2 shows that the fouled values (60 min) lay between 13% and 18%, while the washed values (90 min) were very much the same.

For alumina, which is chemically inert and shows no tendency to aggregate, the BP1, BP2 and BP3 values, with and without soap, mostly lay above 100%. This was to be expected as the alumina, which has a diameter of 1.0 µm, should not have lodged in or stuck to the 0.2 µm pores. Hence one could conclude that RO back pulsing at 140 kPa effectively cleaned non-sticky powders and that there was no need for soap solutions. The mechanism by means of which the back pulsing increases the flux flow rate was not known.

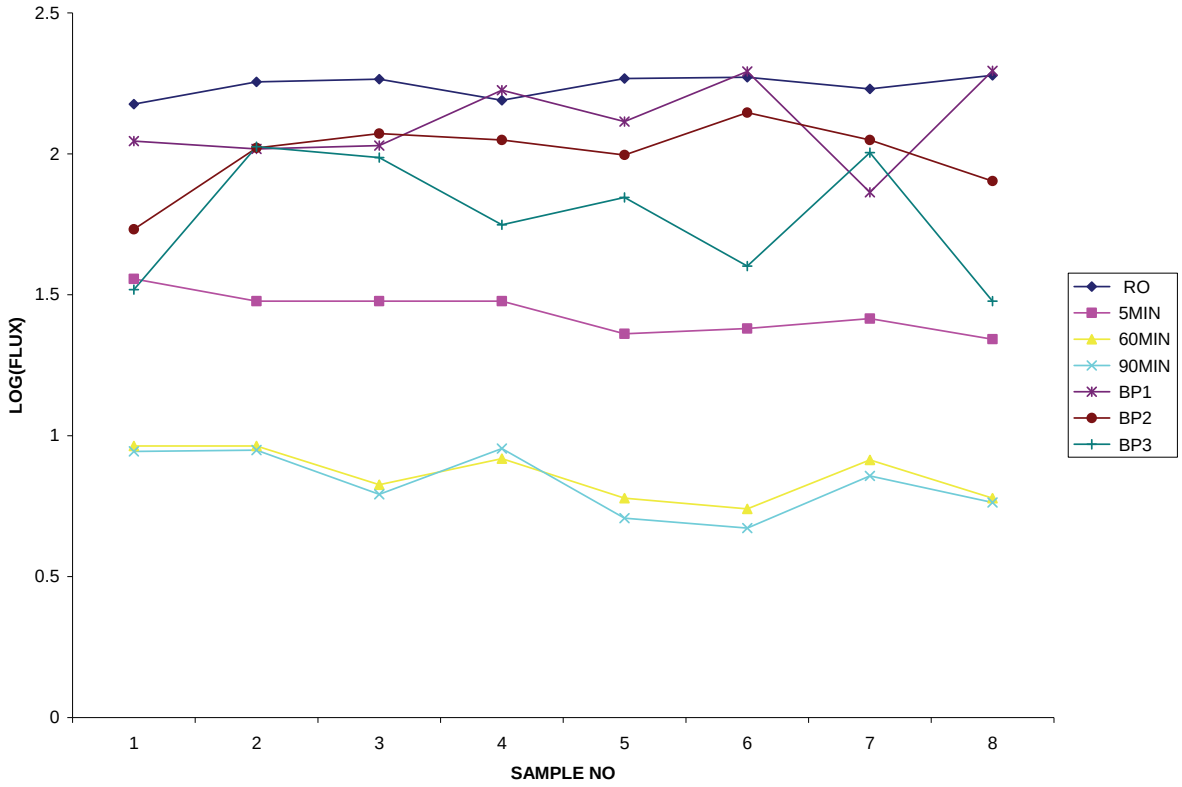

Figure 12. Plot for the PAL 0.45 µm/yeast systems of the flux at the intervals specified in Table 1

B) PAL nylon (0.45) µm membrane with yeast (Figures 8 and 12).

Table 2 shows that when a PAL nylon 0.45 µm membrane was used with yeast as the foulant, the fouled values (60 min) lay between 31 and 61%, while the washed values (90 min) were lower. Yeast, which had a diameter much larger than the alumina, fouled the membrane less effectively than alumina. (SEM results showed that the yeast cells were plugging the pores). For yeast the BP1 value had increased dramatically in all cases. For three RO water pulses the values tended to stay the same or decreased from BP1 to BP3. When the second pulse was a soap solution the same behavior (the flux values from BP1 > BP2 > BP3 being nearly constant or negative) was observed. All the results with and without soap solution were extremely erratic, making a quantitative analysis of the results impossible. However, the final BP3 values showed that there is certainly no dramatic, if any, improvement when soap solutions were used. Note that unlike for the 0.2 µm nylon membrane, the final value BP3 flux value was always well below the clean water value.

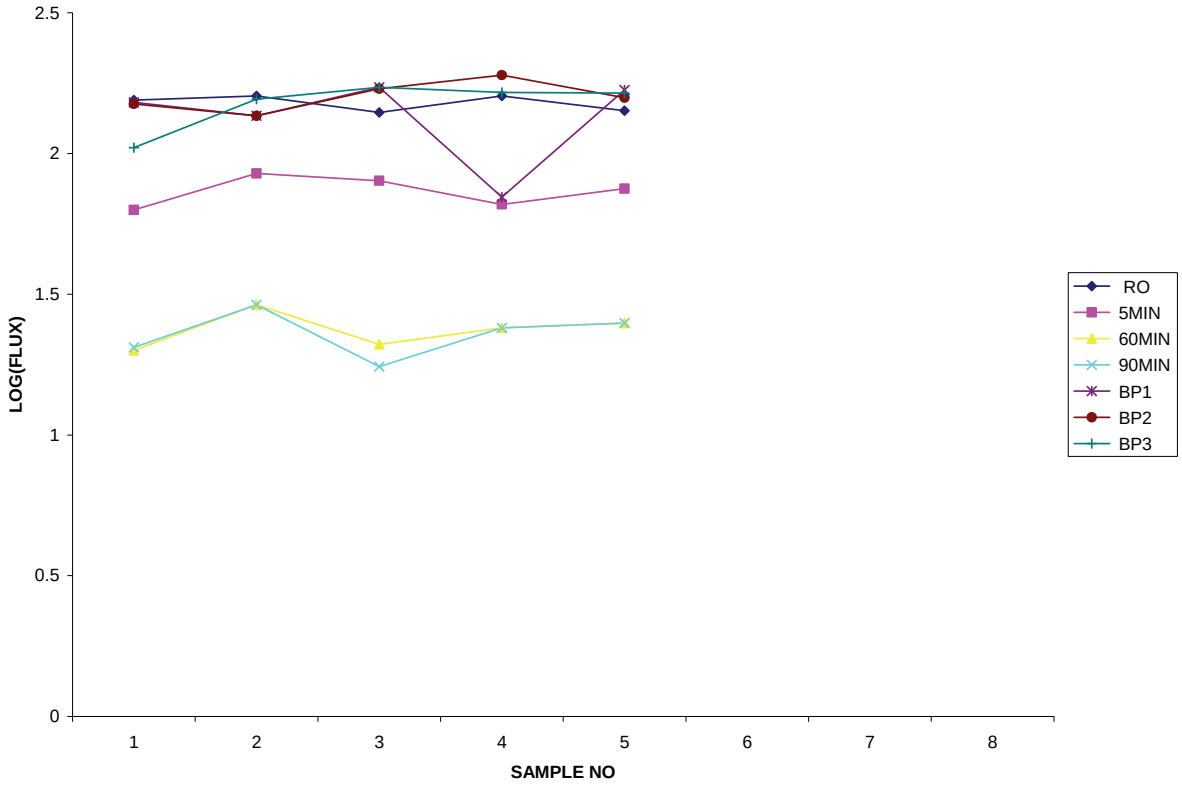

Figure 13. Plot for the GRO/yeast systems of the flux at the intervals specified in Table 1.

C) Polysulphone membrane with yeast (Figures 9 and 13)

A GRO 100 000 MWCO polysulphone membrane was used with yeast as a foulant. Table 2 shows that the fouled flux values lay between 29% and 33% (60 min) and between 28% and 40% for the washed membranes (90 min).

For yeast, using RO for all three back pulses, the BP1, BP2 and BP3 flux values lay close together at about 87%. When the soap solutions S/2, S, 2S and F were used, the final BP3 values were clearly lower than for the all RO water pulse sequence and tended to decrease from BP1 to BP3, as they had done for yeast with the PAL 0.45 µm membrane. This negative slope could be characteristic of when yeast was the foulant. Also it can be seen that a GRO membrane fouled with yeast could not be properly cleaned using three 140 kPa RO back pulses. However, as the combination RO, soap and RO back pulses gave even worse results, it could be concluded that back washing with soap solutions did not improve the cleaning process. Again the reasons for this behavior were not clear, but it is obvious that back pulsing was not as effective at cleaning yeast off membranes as it was for other foulants (alumina and dextrin (see section E below)).

D) Polysulphone membrane with dextrin (Figures10 and 14).

In these experiments, GRO 100 000 MWCO polysulphone membranes were used with dextrin as a foulant. Table 2 shows that the fouled values (60 min) lay between 42% and 52%, while the washed values (90 min) are between 60% and 66%.

For dextrin the flux improved dramatically from the 90min value to BP1 value and usually by only a small amount from BP1 to BP2, irrespective of whether a RO water or soap solution had been used for the second cleaning pulse. The change of the flux from BP1 to BP3 was somewhat erratic, being both positive and negative. The BP3 values often lay above both the RO and the BP2 values, which was similar to the case of alumina on a 0.2 μm nylon membrane. Therefore, from these results it could be concluded that for the GRO-dextrin system, there was no advantage to adding soap to the BP2 back pulse.

The three pulse experiments presented below (see Table 2) were designed to try to clarify the above results by examining the role that colloidal particles played in the fouling.

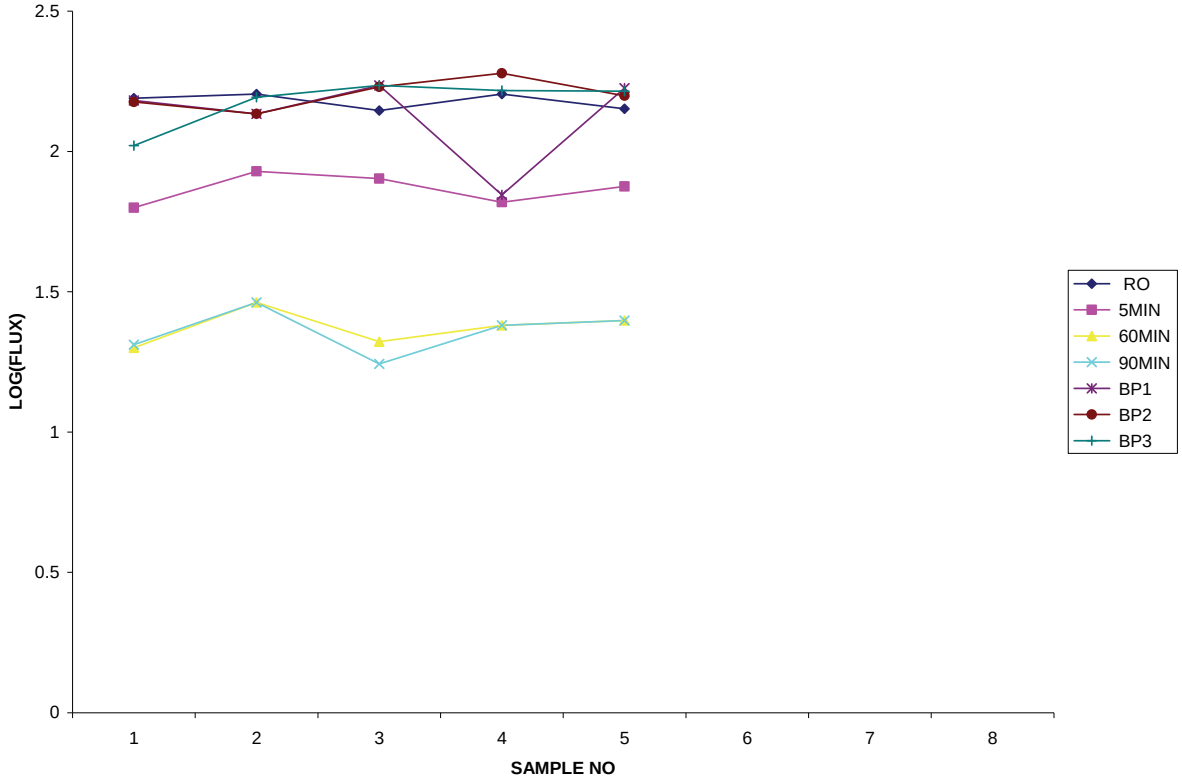

Figure 14. Plot for the GRO/dextrin systems of the flux at the intervals specified in Table 1.

E) Flux flow-time results using "colloidal" foulants (Figures 15-17 and Table 2)

To examine the effect of the colloidal particles associated with the alumina, the alumina colloidal suspension ("milk") was collected from the washing of 1 g of alumina and diluted with 5 ℓ of RO water. Note that the alumina colloids used in this experiment were certainly far more concentrated than in the experiments presented in section 4.2AandB. The results are labeled P2/AM in Table 2 and shown in Figure 15, both of which showed that this suspension was a far more effective foulant (lower 60 minute, BP1, BP2 and BP3 values), than the 1 g of washed alumina per litre, previously used. These results also displayed a continual decrease in flux from BP1 to BP2 to the final BP3 value, indicating that negative

slopes from BP1 to BP3, here and with yeast as foulant (see below) could well have been associated with the colloidal particles. The negative flow may have driven the alumina colloidal particles into the channels in the membrane in an irreversible way (see also section 4.4). The final BP3 value was 5-8 times smaller than for the washed alumina values.

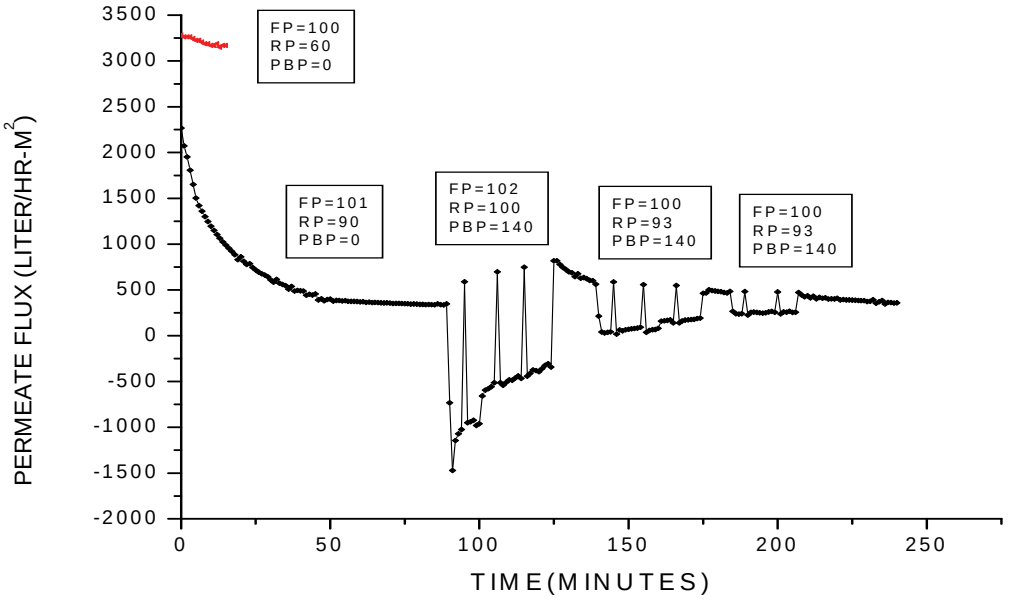

Figure 15. Plot of the flux against the time for the PAL 0.2 µm/alumina (colloidal or "milk") system

In the preceding sub-sections it has been shown that for both the GRO and the PAL membranes yeast was the more effective foulant. As the yeast was live it was washed immediately before use, but could have continued to give off small protein molecules during the experiment. Therefore, the "milk" from the three washes of 1 g of yeast was collected, mixed with 5 ℓ of RO water and used as the foulant for a PAL 0.1 µm (Figure 16) and a GRO membrane (Figure 17). These results also appear in Table 2, as P1/YM and G/YM respectively, and indicate that the colloidal particles which had been washed off the yeast were very effective fouling agents. The P1/YM results in Figure 16, showed the lowest of all the fluxes after BP3 and a strongly decreasing flux from BP1 to BP2 to BP3 (Table 2). This decrease was also a feature of all the P1/Y systems (Table 2), which had already shown low final flux values. However, there are other P1/Y results shown in Table 2 where the flux values are reasonably constant and in these cases the final BP3 flux values were found to be much larger. Therefore one can conclude that the debris from the yeast was probably what caused the drop from BP1 to BP3 where it was observed, but unfortunately the results were not reproducible. It should also be noted that this rapid decrease of the BP1 to BP3 flux values did not occur in any of the other systems given in Table 2.

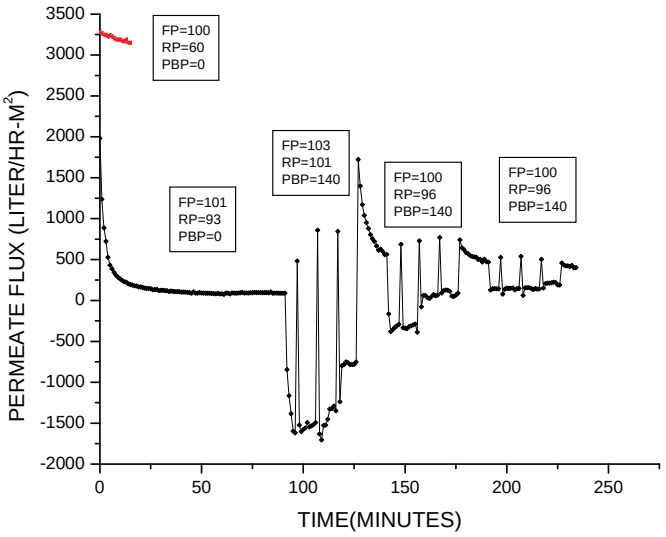

Figure 16. Plot of the flux against the time for the PAL 0.45 µm/yeast (colloidal or "milk") system

In the G/YM system the fouled values were a little higher than for the G/Y systems, but the cleaned G/YM flux value was worse than for the three RO water pulse sequence G/Y experiments. This indicated that yeast colloids had stuck more firmly to the larger pored nylon membranes than to the smaller pored polysulphone membranes.

Note that the amount of colloidal matter generated by the yeast between cleaning and being used in the experiments may have varied, leading to the somewhat irreproducible results. To minimize this possibility the yeast was cleaned shortly before the experiment commenced.

From the above, it can certainly be concluded that these colloidal, or "milk", experiments showed that back pulsing was not very effective at cleaning colloidal sized particles out of either of the PAL nylon membranes, but was somewhat better at cleaning the yeast colloids out of the GRO polysulphone membrane.

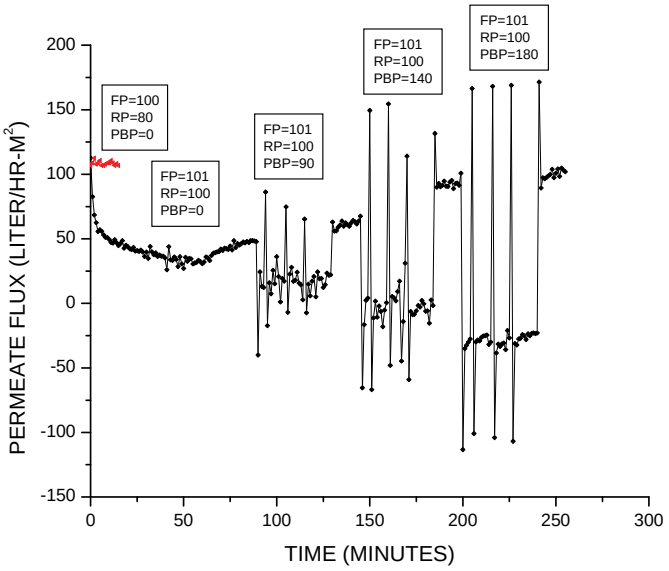

Figure 17. Plot of the flux against the time for the GRO/yeast (colloidal or "milk") system

4.3 Experiments using a two 140 kPa pulse sequence (Table 2 – no figures)

These experiments were carried out to see if back pulsing for the first 30 minutes carried out using a soap solution and not RO water, gave higher flux values after BP1 and/or BP2 than in the previous three pulse experiments. Figures illustrating these results are not presented and the discussion is made based on the results shown at the bottom of Table 2. The experiments were carried out using only the standard SMBS solution, as this soap appeared to have given the best results when a soap solution was used in the three pulse experiments.

The P2/A-S results after both BP1 and BP2 were very similar to those which had been observed in the three pulse experiments.

The P1/Y-S results after both BP1 and BP2 were again very similar to those which had been observed in the three pulse experiments.

The G/Y-S results after both BP1 and BP2 were somewhat better than those which had been observed in the three pulse experiments.

The G/D-S results after both BP1 and BP2 were yet again very similar to those which had been observed in the three pulse experiments.

The conclusion of these experiments was that back pulsing with soap solution first did not improve the overall cleaning obtained by the procedures used earlier in the three pulse experiments.

4.4. Ultrasonic measurements (Figures 18-21 and Table 2)

The thickness of the fouling layer was measured using the apparatus described in the experimental section and in Figures 1 and 3. The UDTR techniques are described in more detail in the final WRC report on WRC project K5/1441. The fouling layer thicknesses, at the end of fouling (60 minutes), are also listed in Table 2.

Figures 18 to 21 show the amplitude of the reflected pulse as a function of the time for the results given in Figures 7 to 10. The 0 minute signal shows the reflections of the signal off the water/new membrane, the internal membrane and the membrane/support interfaces just before fouling begins. The position and shape of the initial reflection depended on the membrane being used, which also affected the reflection from the membrane/support. In all cases, except for dextrin, the signal due to the water/foulant layer was initially (2 to about 10 minutes) only a distortion of the first reflection observed at 0 minutes. After 10 minutes of fouling the new peak became more and more visible and moved to earlier times, indicating the growth of a fouling layer. In all cases, except when dextrin was used as a foulant, these measurements showed the build-up of a fouling layer, several hundred microns thick, after 60 and 90 minutes. The results also showed that the thickness was not influenced much by the washing with RO water (thickness at 60 minutes > 90 minutes). In all cases it could be seen that the first back pulse removed all or most of the fouling layer, which was probably the major cause of the increase in flux after BP1. However, the initial (first) reflection given at 135, 185 and 235 minutes showed that the membrane had been altered; either due to some remnants of the foulant, a distortion of the membrane or both.

The value for the fouling layer thickness was calculated by measuring the difference in time between the reflections from the evolving water/foulant interface and those of the initial water/foulant interface or the membrane/support interface, with due allowance being made for the thickness of the membrane. The actual thickness of the layer was arrived at using these times and the velocity of sound in the media. The identification and interpretation of the reflections, used to measure the layer thickness, required a considerable amount of previous experience and will not be discussed here.

These results are discussed later in conjunction with the results of the SEM work.

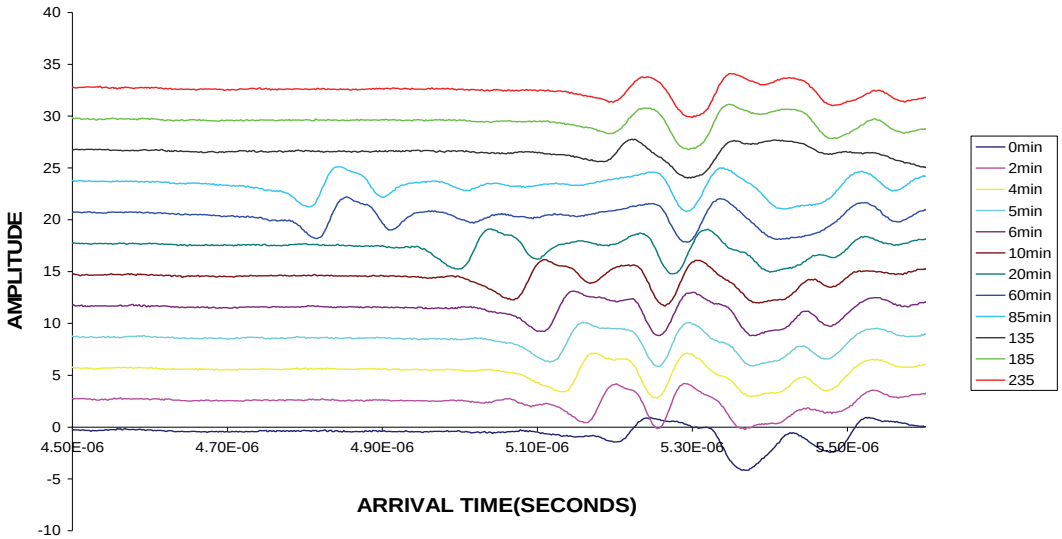

Figure 18. The amplitude of the reflection received at the detector as a function time, for a PAL 0.2 µm/alumina system

Figure 18 shows the amplitude of the reflection received at the detector as a function time, for a PAL 0.2 µm/alumina system. The time intervals shown encompass all the reflections received for the water/film (if a fouling layer was not present, the membrane), film/membrane and membrane/sintered support interfaces.

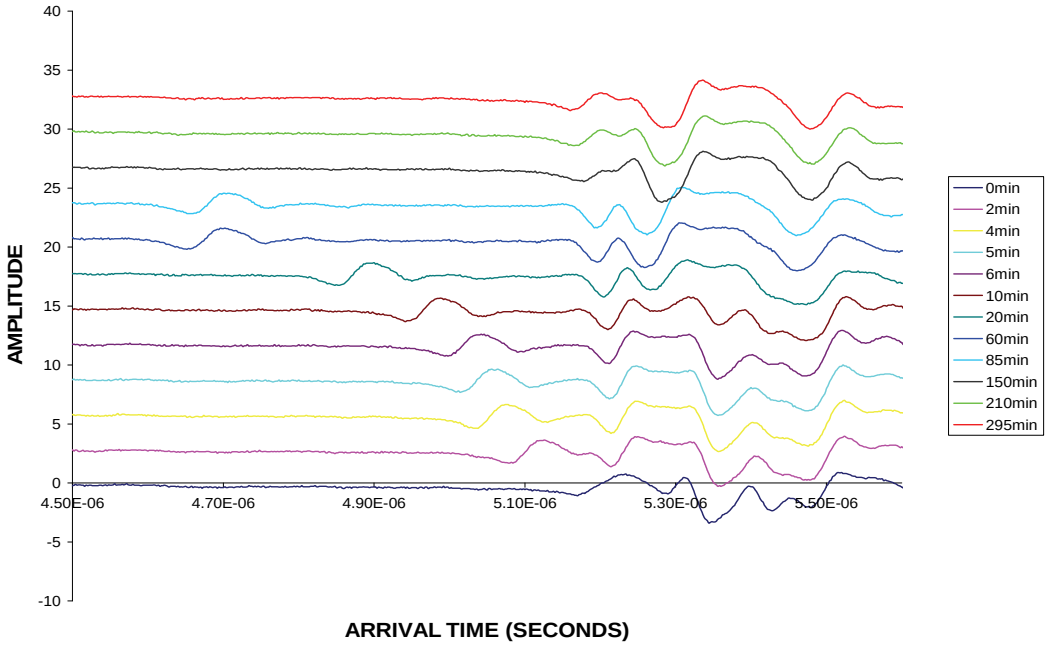

Figure 19. The amplitude of the reflection received at the detector as a function time, for a PAL 0.45 µm/yeast system

Figure 19 shows the amplitude of the reflection received at the detector as a function time, for a PAL 0.45 μm/yeast system. The time intervals at which data were shown encompasses all the reflections received for the water/film (if present if not the membrane), film/membrane and membrane/sintered support interfaces

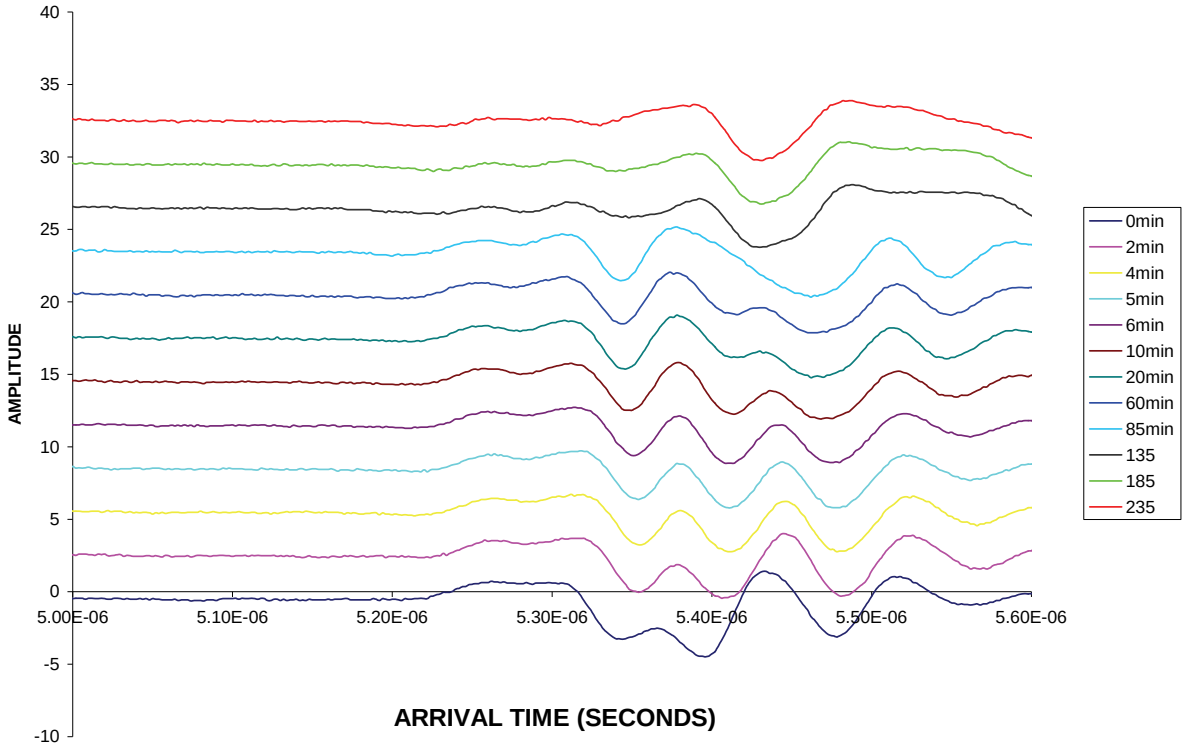

Figure 20. The amplitude of the reflection received at the detector as a function time, for a GRO/yeast system.

Figure 20 shows the amplitude of the reflection received at the detector as a function time, for a GRO/yeast system. The time intervals shown encompass all the reflections received for the water/film (if present if not the membrane), film/membrane and membrane/sintered support interfaces.

4.5. Scanning Electron Microscope Measurements (Figures 22 to 25)

After each experiment the fouled and then cleaned membrane (i.e. after the third back pulse (BP3)) was removed and stored in a glass jar containing 1 g of sodium bisulphate per litre. As samples had to be examined at various times during the experiments, some special additional runs were carried out which terminated after either 15 seconds or 60 minutes. At the end of each of these additional runs the membrane was again stored in the sodium bisulphate solution. The cleaned membranes on systems with a star* in Table 2, column 1 have been examined using SEM. The SEM samples at other times are the result of a special experiment which was terminated at this time.

Note the examination or analysis using a scanning electron microscope and ultrasonic spectroscopy proved the only methods that produced meaningful results and are examined in this report. Other much less successful techniques were infrared spectroscopy and atomic force microscopy and will be mentioned first.

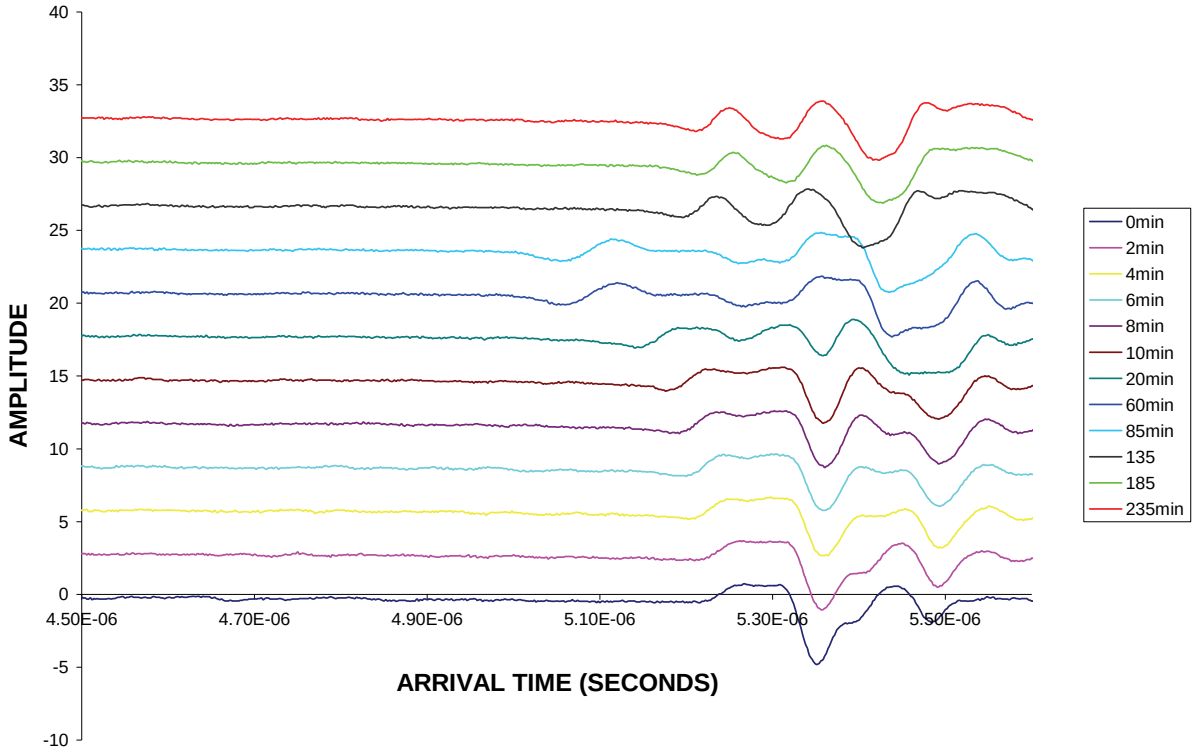

Figure 21. The amplitude of the reflection received at the detector as a function time, for a GRO/dextrin system

[Figure 21 shows the amplitude of the reflection received at the detector as a function time, for a GRO/dextrin system. The time interval shown encompasses all the reflections received for the water/film (if present, if not membrane), film/membrane and membrane/sintered support interfaces.]

New PAL and GRO membranes, fouled samples (60 Minutes) and cleaned samples (235 minutes) were examined, using an infra red spectrometer. However, as the IR signal from the fully fouled membrane was found to be primarily that from the membrane material, with very little, if any, trace of the foulant (this was due to the depth of focus of the spectrometer), no meaningful results could be obtained. Note that pressed pellets of the foulant were measured separately, so that foulant spectra (if present) could have been readily identified. These results showed that to look for traces of the foulant on a fouled and then cleaned membrane was futile.

Atomic Force Microscopy was carried out on the PAL 0.45 μm membrane both in the new, fouled and cleaned state (see SEM image given in Figure 20). The area of the scan was 20×20 μm and the vertical displacement for the new membrane 1.8 to 2.4 μm, for the fouled membrane 1.7 to 3.1 μm and for the cleaned membrane 1.2 to 1.8 μm. The images all had a blurred but brush like appearance, indicating rather spiky or uneven surfaces. In the new and cleaned images the pores could be identified. It was decided not to pursue this line of investigation further as the images yielded no new, useful information compared to the SEM results.

For SEM work a small section (approx. 5×5 mm) was cut from the centre of the wet membrane and allowed to dry. It was then mounted on a SEM stub. Up to 8 stubs could be measured at a time. Images were taken at magnifications of ×500, ×1500 and ×4000. The lower magnifications, which are not shown, were always used to check that the areas examined at ×1500 or ×4000 magnification, were typical of the section of the membrane.

A) PAL 0.2 μm/alumina results: (All images are magnified ×4000)

This system is dealt with first as it gives the most easily understood results. A new membrane surface is shown in Figure 22.1, where the relatively rough pumice-like surface can be seen. Figure 22.2, taken after 15 seconds, where the flux had already dropped considerably, shows no visible pore blocking, but patches of alumina had started to form on the membrane. The mechanism causing the initial drastic reduction in flux is not clear. An image of the fouled surface (layer about 300 μm thick) is given in Figure 22.3 and shows an uneven layer of packed particles. Figure 22.4 shows a membrane that had been cleaned by three RO water pulses. Here, the dark regions are the membrane and the light areas are alumina patches. This image showed that the final surface texture had roughened somewhat from the new surface texture (Figure 22.1), where and the flux value was only slightly lower than the clean water value. The final flux values (BP3) for this system in Table 2 show that this surface was consistent with all previous results.

B) PAL 0.45 μm/yeast results: (All images are magnified 4000 times)

Figure 23.1 shows an image of the new 0.45 μm surface, which is similar to that shown in Figure 22.1, but naturally a bit coarser. After 15 seconds (not shown) the image shows all or most of the pores partially blocked or plugged by yeast cells, which accounted for the initial rapid drop in flux. The fouled surface layer (about 400 μm thick), given in Figure 23.2, showed a dense packing of yeast cells. Figure 23.3 shows a cleaned (three RO water pulses) surface, where the flux was about 66% of the clean water value, which consisted of the odd yeast cell still stuck into the pores plus some unknown debris. Figure 23.4 shows a cleaned membrane where there was still a thin layer of yeast cells on the surface, plus some of the previously mentioned debris. Consistent with this observation was that the flux observed was now only about 20% of the clean water value.

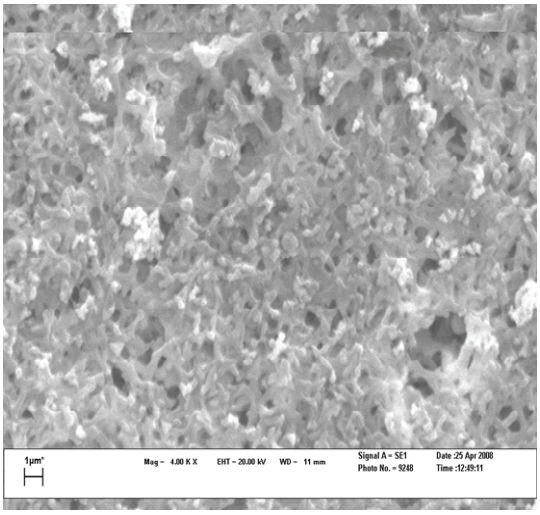

Figure 22.1

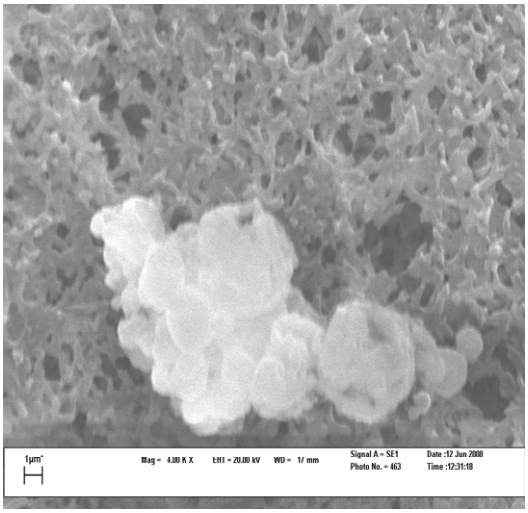

Figure 22.2

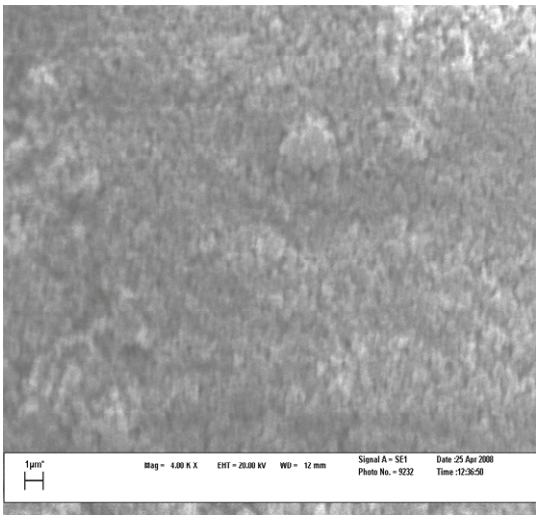

Figure 22.3

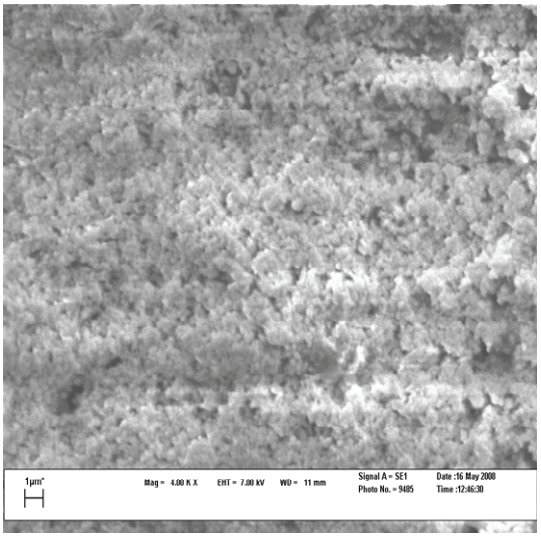

Figure 22.4

Figure 22. SEM images for PAL 0.2 µm/alumina systems

[Figure 22 shows SEM images for PAL 0.2 µm/alumina systems. Figure 22.1 is a new PAL membrane surface. Figure 22.2 shows a similar surface after being fouled for 15 seconds. Figure 22.3 shows a fully fouled (60 minute) surface. Figure 22.4 shows a surface that has been cleaned by three successive RO water pulses.]

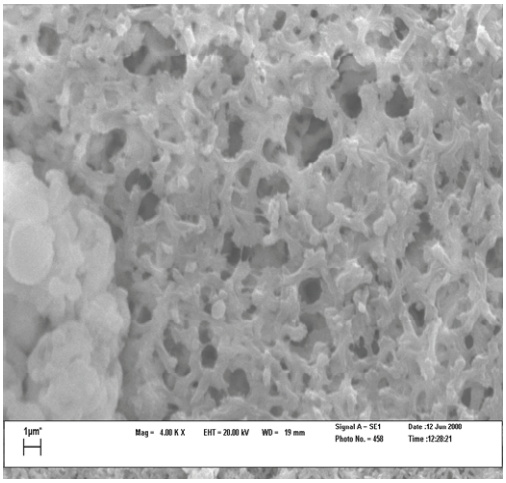

Figure 23.1

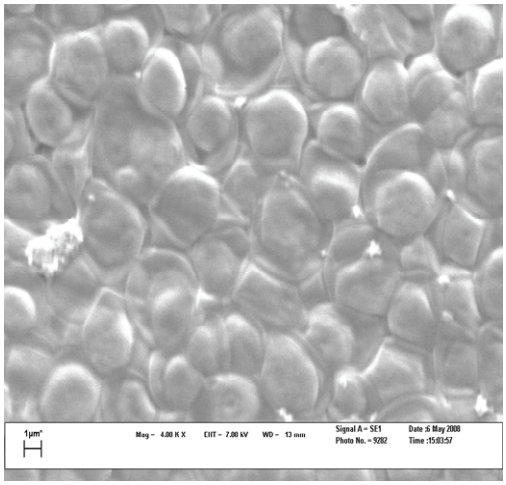

Figure 23.2

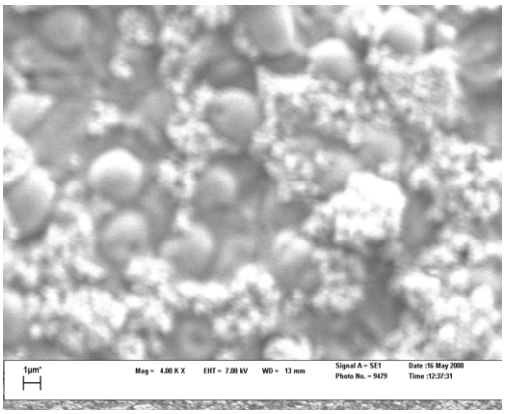

Figure 23.3

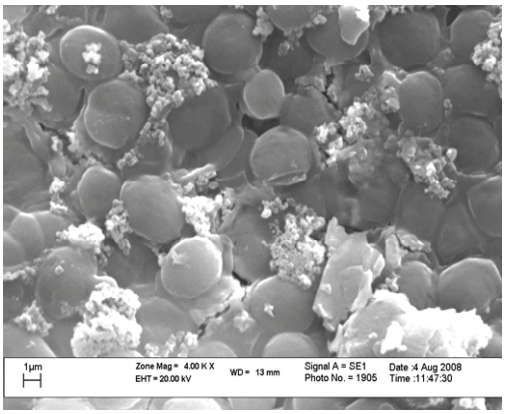

Figure 23.4

Figure 23. SEM images from PAL 0.45 µm/yeast systems

[Figure 23 shows SEM images from PAL 0.45 µm/yeast systems. Figure 23.1 is for a new PAL membrane surface. Figure 23.2 shows the yeast surface after 60 minutes of fouling. Figure 23.3 shows a surface that has been cleaned by three RO water pulses. Figure 22.4 shows a surface that has been cleaned by successive RO water, SMS solution and RO water pulses.]

C) GRO/yeast results: (All images are magnified 4000 times)

Figure 24.1 will show a new surface for a GRO membrane. At 15 seconds (Figure 24.1) one sees the yeast cells clustered around the "invisible" pores, which have been partially blocked by the yeast. Figure 24.2 shows a fouled membrane (60 minute), and that the yeast cells were even more densely clustered around the pores. A layer thickness about 200 µm was obtained from ultrasonic measurements, but the "layer" detected could well have consisted of islands of foulant, higher than 200 µm. It would also appear that the cells have burst. Figure 24.3 is the image of a cleaned (three RO water pulse) surface, which shows large areas of fully cleaned membrane surfaces. This membrane gave a flux of 90% of the clean water

value. Figure 24.4 shows a cleaned (RO water, SMBS, RO water pulses) surface, which shows several areas of packed yeast and as well as the membrane surface. At 500 times magnification (not shown) large areas of clean membrane surrounded by packed yeast could be seen in the image. However, in spite of this structure, the flux for this membrane was still 85% of the clean water value.

D) GRO/dextrin results: (All images are magnified 4000 times)

Figure 25.1 shows a new surface for a GRO membrane. Note the small amount of debris on the surface. As the membrane has a MWCO of 100000, the pores cannot be resolved. The surface does show some texture and the odd bit of debris. The fouled surface, shown in Figure 25.2, appears smooth on this scale but there appears to be a higher density of debris on the surface than in Figure 25.1. The ultrasonic results showed that the dextrin fouling layer was very thin and that it must have had an acoustic impedance similar to that of water, as no clear fouling peak was observed. Hence the thickness of the layer was indeterminable. Figures 25.3 and 24.4 show cleaned (three RO water pulses) surfaces, with fluxes of 9.0 and 15.4 (close to the clean water value) ℓ/hr m^3 respectively. There was obviously less debris on the surface of the cleaned membrane with the higher flux. The origin of this debris was not known and nor was the mechanism by which it appeared (based on this evidence) to have impeded the flux.

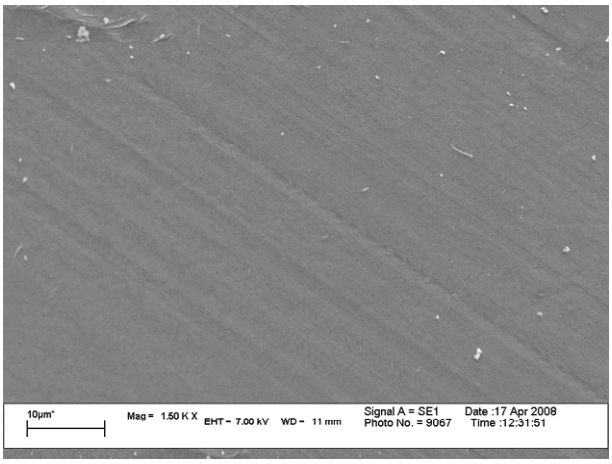

Figure 24.1

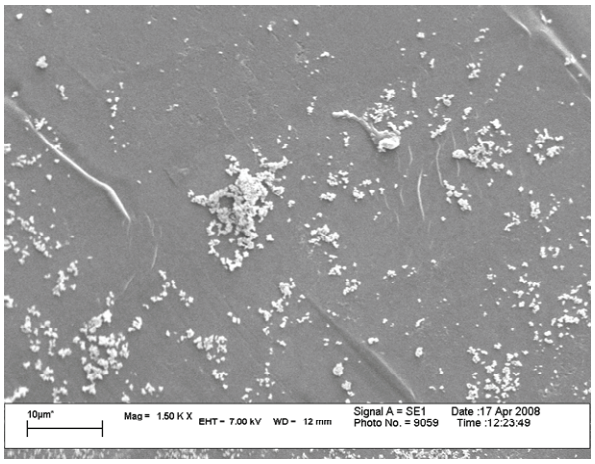

Figure 24.2

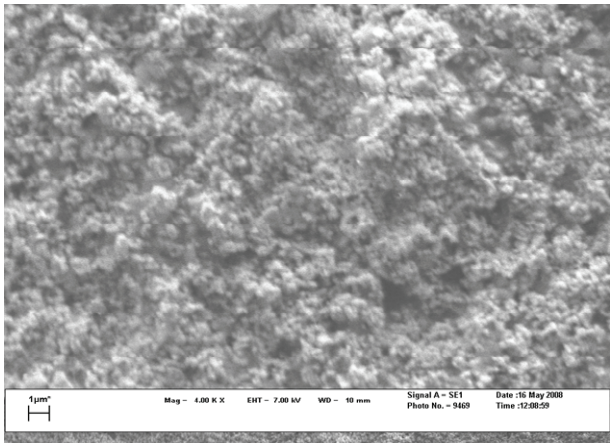

Figure 24.3

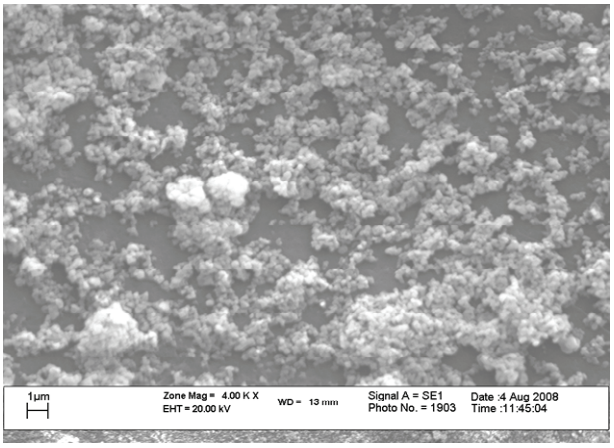

Figure 24.4

Figure 24. SEM images from GRO/yeast systems

[Figure 24 shows SEM images from GRO/yeast systems. Figure 24.1 shows a membrane surface that has been fouled for 15 seconds. Figure 24.2 shows the surface after being fouled for 60 minutes. Figure 24.3 and 24.4 show the surface for two 3 RO water pulse systems which has a somewhat different morphology.]

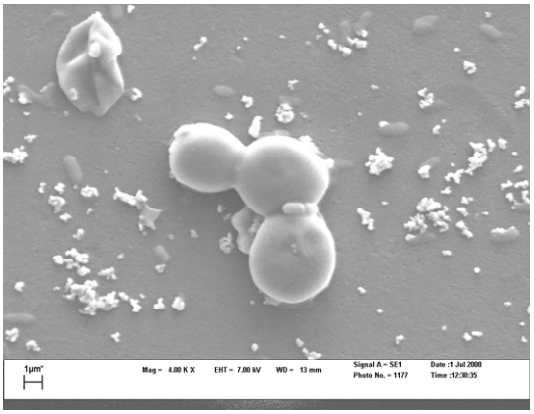

Figure 25.1

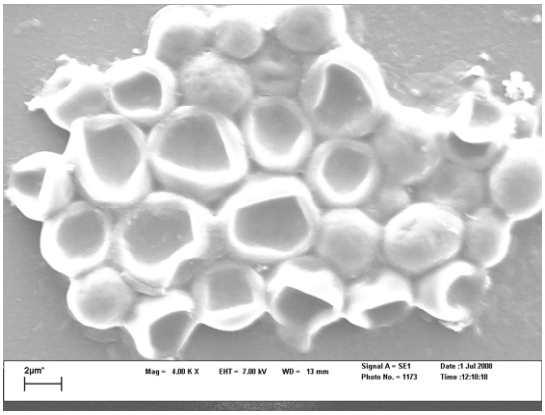

Figure 25.2

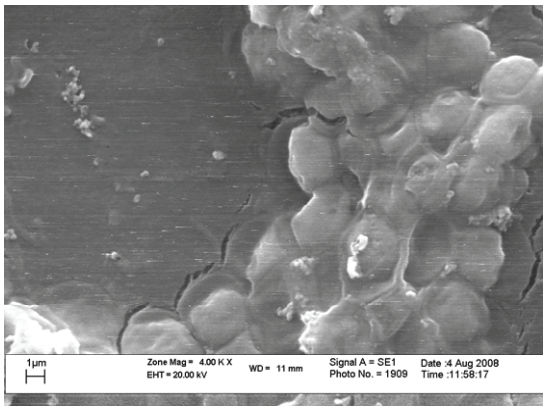

Figure 25.3

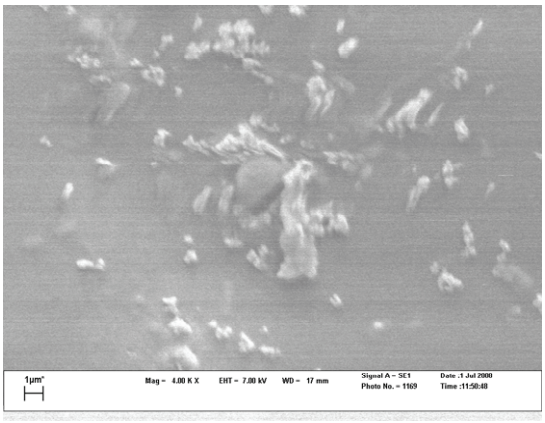

Figure 25.4

Figure 25. SEM images from GRO/dextrin systems

[Figure 25 shows SEM images from GRO/dextrin systems. Figure 25.1 is a new GRO membrane surface. Figure 25.2 shows a surface which has been fouled for 60 minutes. Figures 25.3 and 25.4 surfaces that has been cleaned by three RO water pulses, but which have BP3 flux values of 9.0 and 15.4 ℓ/hr.m^2 respectively.]

4.6 Correlation of the final membrane structure from SEM and US measurements

A) PAL 0.2 µm/alumina:

From Figure 18 it can be seen that the initial peak, at t = 0, which was due to the water/membrane interface and internal reflections from the membrane structure, was almost flat, but slightly negative. The membrane itself was inhomogeneous (as is P1), consisting of two nylon 50 µm layers on either side of a 50 µm woven nylon support, which by itself gave rise to a complex set of reflections, even at t =0. Immediately fouling commences the first peak started to split into a water/foulant and a foulant /membrane peak due to the formation

of the caking layer. This water/foulant peak shifted to earlier times, due to the increasing thickness of the alumina layer, up until 60 minutes. However, after cleaning the first peak structure was almost coincident with the original peak, but now as a split peak instead of the original single, wide peak. The SEM Figure 22.4 shows very little alumina on the cleaned surface, but nevertheless this cleaned surface appears to have split the first reflection.

In the experiment with the alumina colloidal foulant (not shown) a very small 46 μm film was just discernable after 60 or 85 minutes of fouling. However, the originally split membrane peak shape was not much affected by the presence (fouled for 60 minutes) of the thin alumina colloidal layer.

No real information regarding the cleaning of this system was gathered by this analysis but the difference in fouling process for the powder and colloid was re-emphasized.

B) PAL 0.45 μm/yeast

Figure 19 shows a broad double humped membrane peak at t=0, which became flatter during initial fouling, becoming double humped again as the fouling layer thickened. Note that the structure of this membrane is similar to the 0.2 μm one discussed above. The peak due to the yeast layer became clearly visible after 2 minutes and continued to grow up to 405 μm at 60 and 85 minutes. After cleaning a distinct peak at an earlier time than that for the initial membrane (t=0) peak formed, which indicates that some yeast was still clinging to the surface after cleaning. This is confirmed in the SEM image of this surface, shown in Figures 23.3 and 23.4. Note that first (P1/Y) experiment showed the thickest of all yeast layers at 60 minutes, and the lowest final flux of 33 ℓ/hr.m^2, at 235 minutes, which was almost certainly due to the residual yeast layers (see Figure 23.4). The final fluxes for P1/Y 2 and 6 were much higher at 100 ℓ/hr.m^2. The fouling layer at 60 minutes, for these high final flux surfaces was much thinner (Table 2) than for the P1/Y system, as might be expected. An SEM of one of these cleaned surfaces (Figure 23.3) shows scattered imbedded yeast molecules and debris.

C) GRO/yeast

Figure 20 shows a t=0 GRO peak somewhat similar to that seen in Figure 19 for a PAL membrane. [Note this is also a composite membrane as the GRO membrane consists of 60 μm of "membrane" polysulphone on top of a more porous 60 μm polysulphone support layer]. The "peak", due to the ever-thickening yeast layer, was again just visible after 2 minutes. There was a marked change in shape of the membrane peak at 20 minutes, which persisted up to 60 minutes. The final thickness was 199 μm. The cleaned membrane peak at 135 minutes was somewhat similar to that at t=0, but a clear double peak has appeared. There is a more marked first double peak than for the P1/Y system. However, as this type of cleaned peak only occurred when using yeast as the foulant, this type of peak could be due to the remnant yeast observed on the cleaned membranes using SEM.

D) GRO/dextrin

In Figure 21 the water/membrane peak appears to have been barely affected during "fouling" (i.e. up to 60 minutes). This leads one to believe that no substantial dextrin layer was formed, which is confirmed when Figures 21.1 and 21.2 are compared. However, when this UDTR invisible layer was removed by back pulse cleaning, the shape of the initial membrane peaks changed substantially, and a large minimum between the first two peaks was observed. This minimum occurred at a different time from the minima observed for the PI/Y and G/Y systems. The cleaned surfaces, shown in Figures 21.3 and 21.4, correlate with the observed high flux values. In both of these the density of debris on the surface, which presumably does not impede the flux, was found to be considerably higher than on the fouled membrane surface. Either this debris or a distortion of the membrane accounted for the different shapes of the membrane peaks observed after cleaning.

5. SUMMARY AND CONCLUSIONS

The first set of experiments involved cleaning the membrane with successive 90, 140 and 180 kPa pulses, using the permeate water, of 30 minutes each. All these experiments showed that the back pulsing increased the flux rate. The increase was least after the 120 kPa pulse and most after the 180 kPa pulse. At this point the conclusion could be made that back pulsing, with permeate at 180 kPa, could satisfactorily clean the three types of membranes used, of the three foulants listed above. However, as continuous back pulsing at 180 kPa could result in long term damage in spiral wrap membranes, an investigation as to whether, using soap solutions, the same or better cleaning could be obtained at lower back pulse pressures, was made.

This investigation also showed that back pulsing with RO water alone, using the three pulse sequence, can (at least in most of the systems tested in this report) clean the membranes so that the cleaned water flux values (BP3) lie between 70 and >100% of the original clean water (RO) value. The exception to this was P1/Y system, where these values lay between 22% and 58%. From flux-time plots (Figures 7-10), UDTR (Figures 18-21) and SEM (Figures 22-24) measurements (summarized in Table 2) it can be seen that the back pulsing always removed all (or nearly all in the case of the P1/Y system) of the fouling layer but was less effective in removing the particles trapped in the pores. This is supported by measurements made using colloidal suspensions, obtained from the live yeast and alumina. Except for the P1/Y system the BP3 flux values were sometimes equal to, but often much higher than, the flux value after 5 minutes. This indicates that the through membrane back pulsing can only remove some of the "initial" layer. It should be noted that removing the initial layer does not really increase the long-term efficiency of the membrane (element), as the initial layer returns within the first minute (or even seconds) of operation.

The BP1 flux values showed the major removal of the foulant was during the first series of back pulses. This is supported by the UDTR results. In the case of the P1/Y system, the BP1

values indicate that, in some instances, the back pulsing should be carried out for as short a period as is possible. Time prevented this from being investigated further.

From BP1 to BP3 flux values in Table 2 it can be seen that:

a) The change in the flux for the P2/A system was the most irreproducible of all the systems studied.

b) In the P1/Y system the flux usually decreased, sometimes drastically, between the first and third back pulses.

c) The change in flux for the G/Y system was erratic, as sometimes it increased by a small amount but sometimes it decreased by as much as 30%.

d) In the G/D system in all but one case the flux increased, sometimes by about 20%.

The conclusion, based on the above, is that a single back pulse of 30 (or even less) minutes could probably clean most membranes (and possibly elements) sufficiently for them to be reused.

When soap solutions (S, S/2, 2S and F9) are used, the results (Table 2) show that:

a) For the P2/A system the soap solutions increased the final flux, which was surprising as the Alumna particles are the most chemically inert.

b) For the G/D and P1/Y systems no marked improvement could be seen in the rather irreproducible data.

c) For the G/Y system the final flux value could even decrease when soap solutions were used.

None of these results justify using soap solution in conjunction with back pulsing. This is especially true as the soap solutions that would have to be introduced into the plant would later have to be flushed out and disposed of in an environmentally friendly way.

Looking further at all the results, it would appear yeast is the hardest foulant to remove using back pulsing through the membrane, especially for the nylon membrane. The yeast colloid experiment indicates that, if the cleaned yeast continues to make colloidal particles, this is probably the reason why the yeast is harder to clean out of the membrane than the other foulants.

The conclusions from the three (and two) pulse experiments are:

Back pulsing through the membranes, with peak pressures of 140 kPa and more, very effectively cleans the membranes, with the new clean water values usually being in the range of 70 to 100% of the initial values. This shows commercial possibilities and the method could, after further study, be extended to spiral warp and capillary plants.

The main action of the back pulse was to remove all or a significant fraction of the fouling layer. In some cases a remnant of the surface layer could be seen in the SEM image. It is not clear as exactly to what extent the instantaneous flux and the final cleaned value of the flux were influenced by colloidal particles trapped in the pores.

Using soap solutions during the back pulsing never increased (it sometimes decreased) the final cleaned value significantly. Certainly the improvement was never enough to justify the additional trouble and expense of cleaning using soap solutions in conjunction with back pulsing.

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