

# Investigation of Membrane De-clogging Techniques in the Submerged Membrane Filtration Adsorption Hybrid System (SMFAHS)

Paul James Smith,<sup>1</sup> Saravanamuth Vigneswaran,<sup>1\*</sup> Huu Hao Ngo,<sup>1</sup> Roger Ben-Aim,<sup>2</sup> Hung Nguyen,<sup>1</sup>

<sup>1</sup>University of Technology, Sydney, PO Box 123 Broadway NSW 2007 Australia

<sup>2</sup>Institut Nat. Sciences Appliquees, Complexe Scientifique de Rangueil, 31077 Toulouse Cedex 4, France

\*Corresponding Author. Tel: +61-02-9514-2641; Fax: +61-02-9514-2633;

Email: s.vigneswaran@uts.edu.au

*Membrane clogging is a major obstacle to the successful operation of the membrane separation process. A submerged hollow fibre membrane with powdered activated carbon (PAC) adsorption (adsorption-membrane hybrid system) was used for the removal of organics from a synthetic wastewater representative of biologically treated sewage effluent. PAC usage successfully adsorbs the majority of the organics, and then the organic laden PAC is separated by the membrane reducing the direct organic loading to the membrane. However, membrane clogging still occurs.*

*This study involved the development of an automation system and Supervisory Control and Data Acquisition (SCADA) system for performing an investigation and evaluation of three automated de-clogging techniques.*

*The first de-clogging method involved the use of periodic relaxation, whereby permeate production for 12 minutes was periodically stopped for 3 minutes and the shear forces created by the aeration system and the absence of suction pressure during the relaxation period were used to de-clog the membrane. The second de-clogging method involved the use of a series of periodic back flush experiments with varied frequencies and durations to force permeate in the opposite direction out through the membrane pores. The optimal results in terms of de-clogging the membrane were achieved using a 15 second backflush after 15 minutes of permeate production. The third de-clogging method involved the application of an understanding of results of the periodic back flush series of experiments to design an automation system with a new approach to backflushing where an upper limit of a transmembrane pressure (TMP) increase each cycle was used to initiate the backflush. The transmembrane pressure represents the pressure measured across the membrane and it is a vital parameter indicating the degree of fouling of the membrane.*

*A periodic backflush was found to be significantly more effective in terms of increasing the total quantity of wastewater treated than was achieved using periodic relaxation and was investigated in detail during the study.*

*For the periodic backflush, an optimal frequency and duration was determined for treatment of wastewater with a fixed foulant concentration. The new approach to backflushing using more advances in the control system incorporating the TMP increase each cycle resulted in a 40% reduction in the number of backwashes required and was capable of self-optimising operating parameters under an unsteady foulant concentration of wastewater.*

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## KEY WORDS

Adsorption, hollow fibre membrane, powdered activated carbon (PAC), Programmable Logic Controller (PLC), Supervisory Control and Data Acquisition (SCADA), Submerged Membrane Filtration Adsorption Hybrid System (SMFAHS), transmembrane pressure (TMP)

## INTRODUCTION

As a consequence of increasingly stringent standards for wastewater disposal and reuse, various new membrane technologies have emerged. Membrane processes such as reverse osmosis and nano-filtration can remove most of the pollutants, including dissolved organics, but their operational costs are high because of high energy requirements and membrane clogging. Microfiltration and ultra-filtration require low energy and thus are cost effective options. However, they still cannot remove dissolved organic matter due to their relatively larger pore sizes. In a number of water reuse applications, mainly in the late 1990s, ultrafiltration or microfiltration were combined with biologic processes in order to remove the dissolved organic matter (Vigneswaran et al., 2003). When coupled with a biological process, microfiltration or ultrafiltration enable removal of biodegradable dissolved organic matter. Microfiltration used in these hybrid systems can be placed either in the external loop or in submerged configuration. Microfiltration used in the external loop operates at high cross flow velocity which requires a high amount of energy. To reduce the energy requirement,

membrane clogging and to simplify the process, a reactor tank with a submerged membrane is used.

The Submerged Membrane Filtration Adsorption Hybrid System (SMFAHS) is emerging as a highly promising water and wastewater treatment technology (Vigneswaran et al., 1991; Seo et al., 1997; Snoeyink et al., 2000; Kim et al., 2001; and Matsui et al., 2001a, 2001b) that combines addition of powdered activated carbon (PAC) in the reactor tank with the membrane system. In this process, the pollutants (particularly the dissolved organic matter) are first adsorbed onto the PAC, greatly reducing the direct loading of dissolved organic pollutants onto the membrane to reduce clogging and significantly enhance the removal efficiency of the process.

Although PAC usage minimises membrane clogging as the majority of the organic matter present is adsorbed onto the PAC particles rather than the membrane, clogging still occurs during long term operation. This results in increased operating transmembrane pressure (TMP), decreased permeate flux or both and eventually leads to the membrane process being stopped for intensive chemical cleaning of the membrane. Simple and effective de-clogging methods can further minimise the membrane clogging.

This research investigates the use of three de-clogging techniques in reducing long-term membrane clogging to maximize the treated water capacity of the membrane system. The first de-clogging method involved the use of periodic relaxation, whereby permeate production was periodically stopped and the shear forces created by the aeration system and the absence suction pressure during the relaxation period were used to de-clog the membrane. The second de-clogging method involved the use of a series of periodic back flushes, whereby permeate was forced in the opposite direction out through the membrane pores. The third de-clogging method involved the application of the results from the periodic back flush series of experiments to design an automation system where an upper limit of a TMP increase each cycle was used to initiate the backflush.

## THEORETICAL CONCEPTS

The results obtained for different cleaning conditions were compared in terms of productivity.

### 1. Periodic Relaxation

The membrane de-clogging technique of periodic relaxation has the advantage of not using any permeate for backflushing. However, it does not produce any permeate during the period of relaxation. The theoretical productivity of the SMFAHS operating with periodic relaxation can be calculated as follows:

$$P_R \text{ (mL/min)} = \frac{PF_{\text{Initial}} \times T_{\text{Producing}}}{T_{\text{Producing}} + T_{\text{Relaxing}}} \quad (1)$$

where:  $P_R$  = Theoretical productivity of MFAHS operating with periodic relaxation without clogging

$PF_{\text{Initial}}$  = Initial permeate flux of SMFAHS

$T_{\text{Producing}}$  = Period of permeate production cycle

$T_{\text{Relaxing}}$  = Period of relaxation cycle

The permeate produced when the SMFAHS operates in periodic relaxation mode is calculated with a continuous integral of the productivity. Practically, there is progressive membrane clogging which results in a reduction of permeate flux and an increase in TMP during the membrane operation. The cumulative flux can thus be calculated as follows:

$$PP_R \text{ (mL)} = \int_0^t \frac{PF \times T_{\text{Producing}}}{T_{\text{Producing}} + T_{\text{Relaxing}}} \cdot dt \quad (2)$$

where:  $PP_R$  = Cumulative permeate produced with periodic relaxation mode

$PF$  = Real-time permeate flux of SMFAHS during operation (varies with clogging)

$T_{\text{Producing}}$  = Period of permeate production cycle

$T_{\text{Relaxing}}$  = Period of relaxation cycle

Another important measure of productivity incorporates losses due to both the periodic relaxation de-clogging technique and the clogging of the system over time.

For the periodic relaxation operation mode, this is calculated as follows:

$$PR_R \text{ (\%)} = \frac{P_R}{(PF_{\text{Initial}})} \times 100 \quad (3)$$

where:  $PR_R$  = Productivity (%) of the SMFAHS operating with periodic relaxation (expressed as a percentage of the productivity of the system producing permeate continuously with no relaxation period or fouling)

$P_R$  = Productivity of SMFAHS operating with periodic relaxation (during a given cycle)

$PF_{\text{Initial}}$  = Initial permeate flux of SMFAHS before clogging

### 2. Periodic backflush

The membrane de-clogging technique of periodic backflushing has the disadvantage of using permeate for the backflush and not producing any permeate during the backflush. The theoretical productivity of the SMFAHS operating using a periodic backflush can be calculated as follows:

$$P_{BW} \text{ (mL/min)} = \frac{PF_{\text{Initial}} \times T_{\text{Producing}} - BF_{\text{Initial}} \times T_{\text{Backwashing}}}{T_{\text{Producing}} + T_{\text{Backwashing}}} \quad (4)$$

where:  $P_{BW}$  = Theoretical productivity of SMFAHS operating with periodic backflush without clogging

$PF_{Initial}$  = Initial permeate flux of SMFAHS

$BF_{Initial}$  = Initial backflush flux of SMFAHS

$T_{Producing}$  = Period of permeate production cycle

$T_{Backwashing}$  = Period of backflushing cycle

In a practical situation, the permeate flux, however decreases with time as the membrane becomes clogged. Considering the decline in permeate flux with time, the productivity during the membrane operation with the periodic backflush can be calculated as follows:

$$PP_{BW}(mL) = \int_0^t \frac{PF \times T_{Producing} - BF \times T_{Backwashing}}{T_{Producing} + T_{Backwashing}} \cdot dt \quad (5)$$

where:  $PP_{BW}$  = Cumulative permeate produced with periodic backflush

$PF$  = Real-time permeate flux of SMFAHS during operation (varies with clogging)

$BF$  = Backflush water flux of SMFAHS

$T_{Producing}$  = Period of permeate production cycle

$T_{Backwashing}$  = Period of backflushing cycle

For the periodic backflush operation mode, the productivity incorporating losses due to both the periodic backflush de-clogging technique and the clogging of the system over time is calculated as follows:

$$PR_{BW}(\%) = \frac{P_{BW}}{(PF_{Initial})} \times 100 \quad (6)$$

where:  $PR_{BW}$  = Productivity (%) of the SMFAHS operating with a periodic backwash (expressed as a percentage of the productivity of the system producing permeate continuously with no backwash or fouling)

$P_{BW}$  = Productivity of SMFAHS operating with periodic backflush (during a given cycle)

$PF_{Initial}$  = Initial permeate flux of SMFAHS before clogging

Even during the operation of the SMFAHS with either periodic backflush or periodic relaxation, the membrane becomes clogged and the permeate flux begins to decline leading to a reduction in productivity from the theoretical value. The reduction in productivity can be minimised by optimising the de-clogging conditions that allow minimal membrane clogging. The membrane clogging is indicated by the continuous TMP drop monitored by the system.

## EXPERIMENTAL

The schematic diagram of the SMFAHS experimental set-up is shown in Figure 1.

Wastewater was pumped into the reactor tank using a feed pump and reed switches to control the level and volume in the tank. A predetermined amount of PAC was initially added into the tank to adsorb the dissolved organic substances, which was subsequently separated by the membrane filtration imposed by the suction pump. No further addition of PAC was made during the experiment. A pressure gauge was used to measure the transmembrane pressure of the hybrid system. An air diffuser was used to maintain the PAC in suspension.

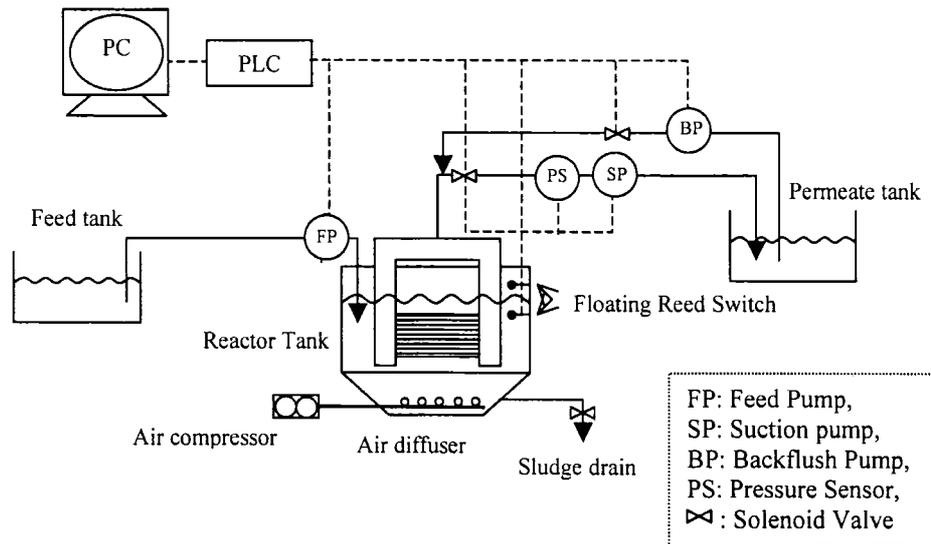


Figure 1. Experimental set-up of the SMFAHS.

This research involved the development of an automation system and a graphical operator interface which enabled backflush optimization, productivity maximization and accurate recording of project data for the SMFAHS. The automation system was developed through the programming and commissioning of a Programmable Controller. The operator interface was based on a Supervisory Control and Data Acquisition (SCADA) system that was programmed to communicate directly with the Programmable Controller.

Using the automation system, periodic backflushing and periodic relaxation methods were employed to de-clog the membrane system. The relative merits of these systems were studied in terms of achieved productivity. Periodic backflushing involved reversing the direction of permeate flow at twice the rate used during the membrane filtration to force permeate out through the membrane and remove foulants from the membrane surface and pores. Periodic relaxation involved periodically stopping permeate production, allowing the shear forces of

the aeration diffuser to remove foulants from the membrane surface.

Initially, periodic relaxation was investigated. The second series of experiments involved varying the backflush frequencies and durations. For all experiments, the ratio of backflushing frequency to backflushing duration was maintained at 60:1. Finally, a more advanced backflush was trialed with the use of knowledge of the TMP drop increase attained from the periodic backflush study as an initiator for the backflush cycle.

Table 1 shows the constituents of the synthetic wastewater used in this study. The synthetic wastewater consists of 12 organic and inorganic with similar properties to biologically treated wastewater.

Table 2 shows the physical and chemical properties of the hollow fiber membrane used in this study.

Table 3 shows the characteristics of the Powder Activated Carbon (PAC) used in this study.

**Table 1. Constituents of the synthetic wastewater used.**

COMPOUNDS	Weight (mg/L)	COMPOUNDS	Weight (mg/L)
Beef extract	1.8	Sodium lauryle sulphate	0.94
Peptone	2.7	Acacia gum powder	4.7
Humic acid	4.2	Arabic acid (polysaccharide)	5
Tannic acid	4.2	(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	7.1
Sodium lignin sulfonate	2.4	K <sub>2</sub> HPO <sub>4</sub>	7
NH <sub>4</sub> HCO <sub>3</sub>	19.8	MgSO <sub>4</sub> ·3H <sub>2</sub> O	0.71

**Table 2. Physical and chemical properties of the membrane.**

Properties	Hollow fibre membrane
Total surface area (m <sup>2</sup> ), (320 fibres with 12cm length)	0.05
Pore size (µm)	0.1
Material	Polyethylene
Inner diameter (mm)	0.27
Outer diameter (mm)	0.41

**Table 3. Characteristics of the PAC.**

Specification	PAC-WB
Iodine number (mg/g min)	900
Ash content (%)	6 max.
Moisture content (%)	5 max.
Bulk density (kg/m <sup>3</sup> )	290-390
Surface area (m <sup>2</sup> /g)	882
Nominal size	80% min finer than 75 micron
Type	Wood based
Mean pore diameter (Å)	30.61
Micropore volume (cc/g)	0.34
Mean diameter (µm)	19.71
Product code	MD3545WB powder
Adsorption capacity g(TOC)/g(PAC)	0.00567*

## RESULTS AND DISCUSSION

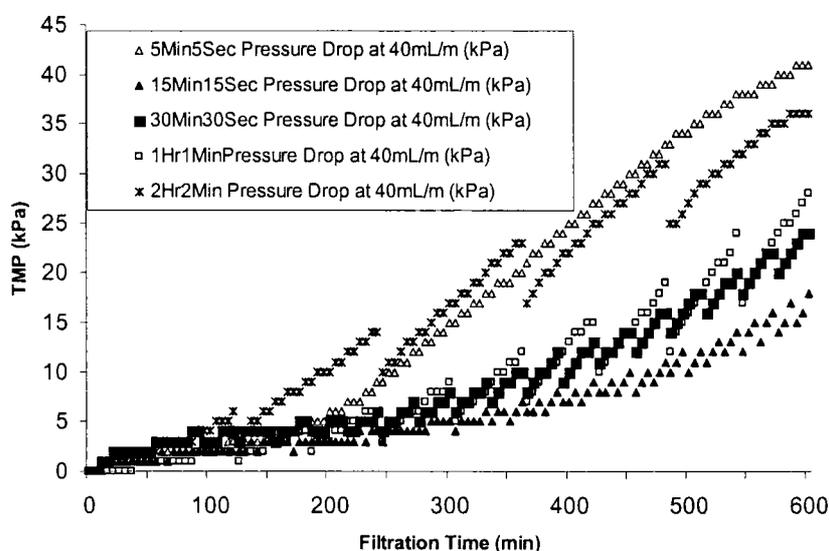
### 1. Time based periodic backflushing

The effect of backflush frequency and backflush duration was studied. Table 4 shows the parameters used in each of the experiments.

Although the periodic backflush reduced the TMP increase rate and thus the membrane clogging, it could not be completely eliminated. Figure 2 shows the results of the five experiments outlined in Table 4.

**Table 4. Backflushing parameters used in the experiments.**

Experiment Number	Backflush Duration (sec)	Backflush Frequency (min)	Backflush Flux (mL/min)
1	5	5	80
2	15	15	80
3	30	30	80
4	60	60	80
5	120	120	80



**Figure 2. TMP profiles with operation time for different backflushing conditions. (Initial Permeate Flux = 40 mL/min)**

Results from the experiment indicate that a backflush duration of 2 minutes at a frequency of 2 hours was insufficient to control the increase of TMP. The backflush duration of 5 seconds was also found to be insufficient even though the membrane was backflushed every 5 minutes. The TMP increase was minimal when the membrane was backflushed for 15 seconds every 15 minutes of filtration time.

From a model developed for the relationship between permeate flux and TMP, the resulting cumulative permeate production and percentage productivity data with time are shown in Figures 3 and 4.

Figure 3 shows the relationship between permeate production and time. The theoretical maximum productivity

curve shows the achievable amount of permeate production with losses only from the backflushing and not from clogging. In real operating conditions, the membrane begins to clog, resulting in a lower permeate flux and deviation from the theoretical productivity curve. The amount of deviation from the theoretical productivity curve is inversely related to the TMP of the membrane which is caused by clogging. By selecting optimal backflushing parameters, this clogging can be minimised allowing the system to operate closer to the theoretical productivity. As well as the productivity improvements attainable using optimal backflushing parameters, the membrane can have a longer life.

Figure 4 shows the deviation of the productivity for the various backflushing parameters.

If clogging was totally alleviated, the system could operate continuously at 95.1% productivity (with a 4.9% reduction due to losses involved with backflushing). However, membrane clogging cannot be avoided and the productivity diminishes. By choosing the optimal backflush conditions, the reduction in productivity can be minimised.

From the preliminary experimental investigation, the optimal backflushing condition was found to be a 15 second backflush after every 15 minutes of filtration. The pressure drop during each cycle was approximately 1.5kPa.

Thus a detailed experiment was conducted using a predetermined value of TMP as the controlling parameter (i.e. as soon as the TMP dropped by 1.5kPa during each filtration cycle, the backflush was started). After a backflush cycle, the permeate pump operates and the TMP begins to stabilize. At this point, the control system logs a TMP reading. The TMP is then recorded and continuously compared to a value 1.5kPa below this recorded TMP reading. Once this value is reached, the backflush is initiated.

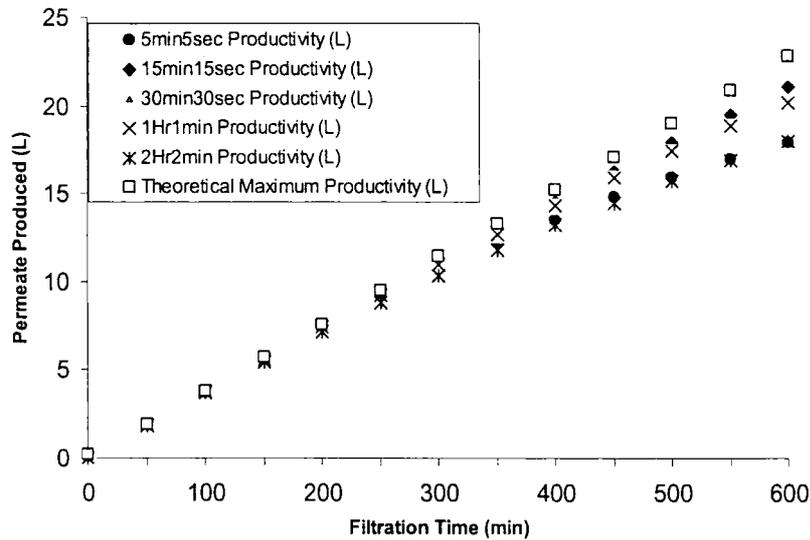


Figure 3 Cumulative permeate production versus filtration time.

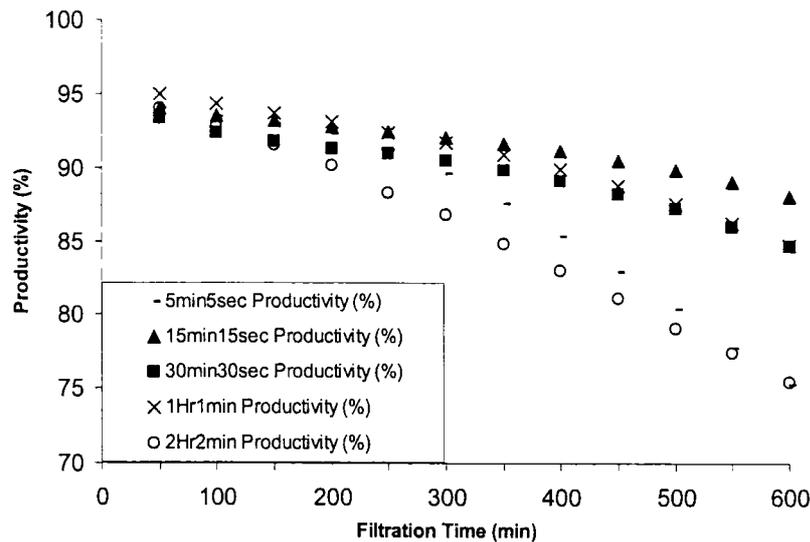


Figure 4. Productivity (%) versus Time.

## 2. Comparison of the three methods

A series of 20 hour experiments were then performed with different modes of cleaning (the optimal timed periodic backflush condition, the TMP based pressure initiation backflush and periodic relaxation).

Over a 20-hour period, TMP based pressure initiation backflush method resulted in a similar long-term pressure drop to the optimal timed periodic backflush, but required less frequent backflashes leading to a further improvement in productivity. This also has the benefit of automatically finding the optimal backflushing parameters for wastewater varying in influent concentration. The backflush was found to be much more successful than relaxation in terms of productivity and clogging.

Figure 5 shows the results of TMP for the optimal fixed time backflush, periodic relaxation and the TMP based pressure initiation backflush with time.

Although there can be no theoretical measure of the maximum productivity from using a TMP based pressure initiation backflush, it resulted in a 40% reduction in the number of backflashes required and more permeate production. The success of these results is due to the backflush occurring when required (TMP drop = 1.5 kPa) rather than at a fixed time when it may not be required.

Figure 6 shows the deviation of the productivity for the both the timed backflush and the periodic relaxation methods of cleaning.

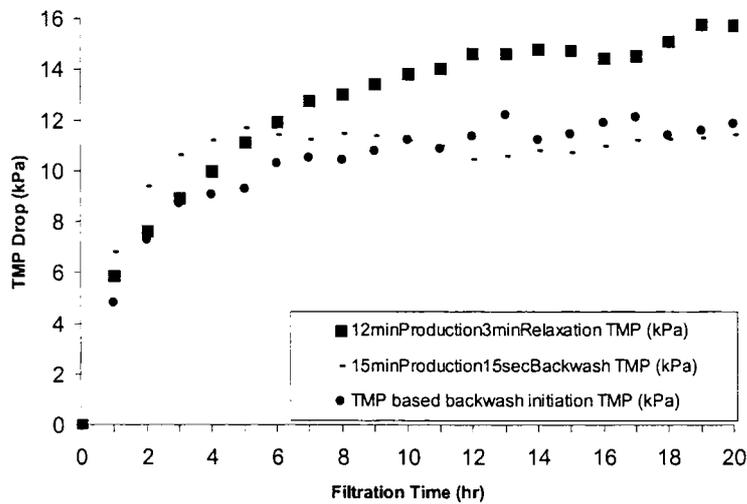


Figure 5. TMP profiles versus time for different de-logging methods.

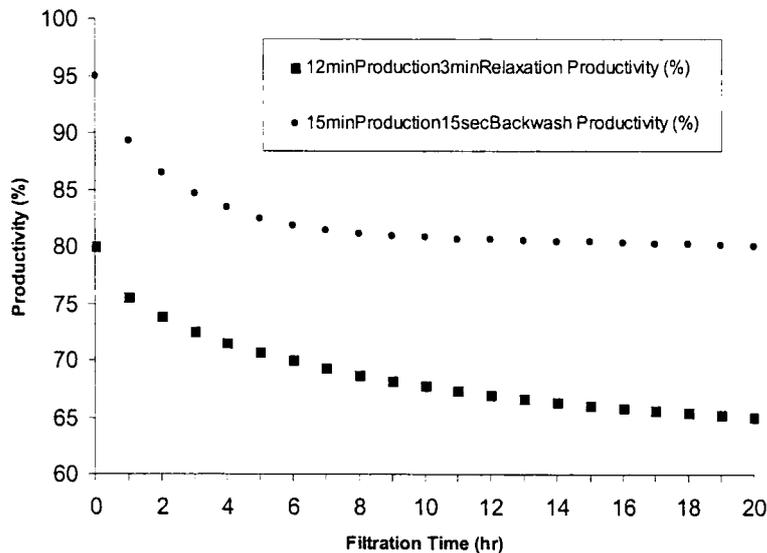


Figure 6 Productivity (%) versus operation time.

As can be seen in Figure 6, the theoretical productivity for timed backflushing is 95.1% and for periodic relaxation it is 80%. During operation, the actual productivity for periodic relaxation decreased at a faster rate than that of periodic backflushing. This indicates that attempting to operate the system using periodic relaxation with an increase in the theoretical productivity through either increasing the permeate production period or

decreasing the relaxation period would be unsustainable as clogging would erode any productivity gains.

Figure 7 shows the cumulative permeate production versus TMP drop for the various de-clogging conditions.

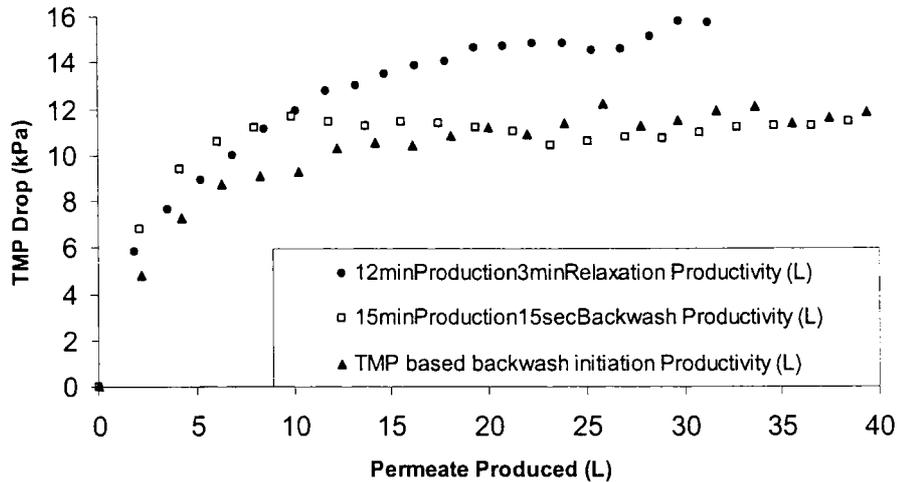


Figure 7 Cumulative permeate production versus TMP Drop for different de-clogging conditions.

Severe membrane clogging is indicated by a TMP drop of over 50 kPa and once reached requires the membrane to undergo an intensive physical and chemical clean. These figures indicate that optimized cleaning methods and parameters can effectively increase the total quantity of wastewater treated before the membrane requires an intensive physical and chemical clean.

## CONCLUSIONS

- Periodic backflushing was found to effectively reduce reversible clogging on the membrane surface allowing a high permeate flux rate to be maintained;
- An optimal frequency for the periodic backflushing exists for this fixed foulant concentration of wastewater;
- The optimal frequency for the periodic backflushing cannot be fixed in case of unsteady foulant concentrations of wastewater;
- A periodic backflush was significantly more effective in terms of increasing the total quantity of wastewater treated than was achieved using periodic relaxation;
- The use of the pressure based backflush system enables the SMFAHS to operate effectively under unsteady concentrations of wastewater as a backflush is only initiated when required rather than at fixed time intervals for a periodic backflush system; and

- The productivity of the SMFAHS is comprised of an initial theoretical limit of productivity that begins to decrease as clogging occurs.

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