

EXECUTIVE SUMMARY

Introduction

Membrane fouling is universally accepted as one of the most critical problems limiting the wider application of membranes in liquid separations. The development and utilisation of a suitable non-invasive technique for the on-line monitoring of fouling in industrial and laboratory applications may enable the effectiveness of fouling remediation and cleaning strategies to be quantified.

The present research project was born from the words of Dr Irwing Murdoch, editor of *Water Desalination and Re-use* and chairman of a task group on membranes who, in 1999, stated that there was a need to revisit problems that limit the reverse osmosis (RO) field. In the project reported on here this was approached by using the tremendous recent gains made in available analytical equipment, including high-frequency digital and computer techniques. This was encouraged and supported by Professors Kranz and Greenberg, then of the Separation Centre, University of Colorado, USA, who had already two or three years experience in ultrasonic techniques. In this research project (#1166) we investigated the use of ultrasonics to detect and monitor in different membrane systems and in different configurations.

Through an initial programme funded by the WRC (#930) in which the use of ultrasonic waves was investigated it was proved that, using ultrasonics, fouling on RO membranes could be seen. This opened a number of doors to this investigation and gave rise to many of the aims.

Objectives

The overall objective of this research is to develop ultrasonic time-domain reflectometry (UTDR) for its use as an analytical tool for the real-time study of inorganic, organic and protein fouling of various types of membranes, including nylon, polysulfone (PSU) and polyethersulfone (PESU), and modules, including flat-sheet and tubular types. Different separation systems including microfiltration (MF), ultrafiltration (UF) and reverse osmosis (RO), flat-sheet and tubular modules, and suitable ultrasonic probes were used in this study.

Specific objectives were subsequently the following:

- In-situ measurement of particle deposition and removal in microfiltration
- Cleaning of microfiltration membranes by ultrasonics
- Determining cake layer deposition, its growth and compressibility, and removal during microfiltration

- Direct monitoring of membrane fouling and cleaning during ultrafiltration
- Measurement of protein fouling in tubular ultrafiltration membranes
- Visualisation and monitoring of inorganic fouling and cleaning of reverse osmosis membranes
- Generation of sufficient data to be able to propose and patent a modified plugging index meter.

Methodology

The overall aim of this project was to develop ultrasonic time-domain (UTD) and amplitude-domain reflectometry (UTDR) as a real-time visualisation technique for monitoring fouling in various pressure-driven membrane separations.

This was approached in the following way:

Consideration was first to be given to various designs for the development of the UTDR technique and suitable test cells. The latter should allow the simultaneous pressure-driven separation and monitoring of fouling. Systems for UTDR measurements and ultrasonic cleaning were set up and used. Flat-sheet and tubular membrane cells were designed and used. A mathematical model was to be developed and used to describe the process of ultrasonic testing, as related to the deposition of fouling in complex membrane environments.

UTDR was first to be used for the non-invasive, *in situ* measurement of particle deposition during microfiltration and, pending successful detection, particle removal, using flat-sheet nylon membranes. Scanning electron microscopy was to be used to confirm UTDR results. Further investigations into the cake-layer deposition, its growth on a membrane surface, and its compressibility were to be considered. This was then to be followed by similar investigations with polysulphone (PS) ultrafiltration membranes. Protein fouling of tubular polyethersulphone membranes (PES) was also to be considered. Finally, UTDR was to be used to monitor fouling and cleaning of reverse osmosis membranes

Results and Conclusions

This report describes the developments and successes achieved in both proving and understanding ultrasonic monitoring of membrane fouling. The overall achievement of this investigation was the development of ultrasonic time-domain (UTD) and amplitude-domain reflectometry (UTDR) as a real-time visualisation technique for fouling monitoring in various pressure-driven membrane separations. It was determined that UTDR can be used successfully

to monitor membrane fouling in MF, UF and RO with different membranes, foulants and configuration modules.

Specific results and conclusions, in the following areas, were as follows.

Microfiltration (MF)

- Ultrasonic response signals were obtained for kaolin particle deposition and subsequent cake layer formation on a nylon membrane surface during microfiltration (MF). A cake layer echo appeared, increased in amplitude, and moved in time due to layer thickening as fouling proceeded in MF.
- An ultrasonic cleaning technique was successfully used for cleaning membrane fouling in MF. It was established that ultrasound associated with forward flushing is a new and effective method for cleaning fouled MF membranes. Analysis by scanning electron microscopy (SEM) indicated that ultrasound with flushing was able to remove fouling layers from a membrane surface and restore the original structure of the membrane surface. The cleaning efficiency obtained by ultrasound with forward flushing was as high as 97.8%.
- UTDR was successfully used to measure the rate of cake layer formation at different flow rates. UTDR was also capable of detecting subtle changes such as the stop and restart operation.
- UTDR was successfully used for monitoring membrane cleaning and evaluating the cleaning effectiveness of various cleaning methods, namely: forward flushing, ultrasonic cleaning and ultrasound with flushing. Results showed that a cake-layer echo decreased after flushing and disappeared after ultrasonic irradiation in MF. UTDR results showed that ultrasound associated with flushing was the most effective cleaning method for cleaning membranes fouled by paper mill effluent.
- UTDR proved to be very useful method for the investigation of fouling mechanisms. The compressibility of the cake layer contributed to the flux decline during fouling. UTDR proved that increasing the flow rate during actual operation could not reduce fouling in MF carried out with paper mill effluent because a denser cake layer formed on the membrane surface at a high flow rate than it did at a low flow rate. An increase in flux could be seen when fouling operation was stopped and restarted. This is because interrupting the fouling experiment resulted in flow destabilisation and relaxation of the fouling layer. UTDR showed an increase in the thickness of the fouling layer and a decrease in the density of the cake layer.

Ultrafiltration (UF)

- In ultrafiltration (UF), an asymmetric polysulphone (PSU) membrane and its compaction could be detected by UTDR. Results showed a good correspondence between the UTDR signal response and the development of the fouling layer on the membrane surface in UF.
- The dynamic processes of deposition and growth of a fouling layer was confirmed by *in-situ* ultrasonic response signals. The sensitivity of ultrasonic responses revealed the coverage of a membrane surface with a fouling layer. UTDR results corroborated the flux measurements and SEM analyses.
- UTDR could also distinguish and recognise various response signals from ultrasonic measurements in a tubular cell. Protein fouling was successfully detected by UTDR in a tubular PESU UF membrane. Changes in the amplitude and time domain of the observed signals provided useful quantitative information about adsorption, deposition and gelation of BSA on the membrane. The thickness of the BSA gel layer was obtained. UTDR can provide an approach to understand protein-fouling behavior related to flux decline.

Reverse Osmosis (RO)

- UTDR was successfully used for the RO of saline water, calcium sulphate streams (such as in mining), real pulp and paper effluent and, more recently, biological systems, and for flat sheets and tubular membranes.
- The application of the UTDR technique is feasible for the non-destructive, in-situ visualisation of membrane fouling and cleaning in reverse osmosis (RO) modules. Results showed that the UTDR technique could effectively detect fouling-layer initiation, its growth on the membrane and its removal, in real-time. The acoustic signal response corresponded well with the membrane's flux-decline behaviour. The UTDR technique was capable of detecting subtle changes on a membrane surface and distinguishing between dead-end and cross-flow modes of fouling growth. This could assist in understanding the mechanism of membrane fouling.

A secondary fouling echo was obtained (the primary echo comes from the membrane). The second echo amplitude should be seen as an indication on the state of the fouling layer. The denser the fouling layer, the better the reflection, and thus the larger amplitude that is seen. The fact that the UTDR technique can monitor the removal of a fouling layer and membrane cleaning makes it a very suitable tool for studying the effectiveness of various cleaning techniques.

- The ultrasonic unit that evolved comprises a suitably programmed microprocessor and display panel, and can be used to compare reference and test signals to produce a differential signal (a fouling layer echo). A differential signal indicates the state and progress of a fouling layer on a membrane surface during actual operations. Both amplitude and arrival time of differential signals as a function of operation time provide useful quantitative information, i.e. changes in thickness and density of a fouling layer on the fouling processes. The resolution exceeds the theoretical limit by a considerable margin when using the differential signals.
- A predictive modelling program - ultrasonic reflection modelling (URM) - was developed to understand the processes of ultrasonic testing related to the deposition of fouling layers on membrane surfaces. The mathematical modelling could substantiate changes in the density and thickness of the cake layer. Deposit resistance is not only related to the layer thickness but also the cake density (compressibility). The predicted results of fouling layer deposition were in good agreement with the actual measurements obtained in MF and UF.
- Sufficient data were created to be able to propose and patent a modified plugging index (PI) meter. (PI meters are the basis of knowing the fouling potential of any feed water, and of studying the efficiency of any pretreatment stream. Therefore a PI meter that can visualise, in addition to its normal functions, has an as yet unexplored market need. Coupled to the fact that the PI meter can be used as a slave unit attached to any bank of membranes, and using equivalent membranes in the slave unit, direct monitoring of the bank is a capability. Further, since it gives an electronic signal it can be used for control of the process and defouling.)

Recommendations

The research has clearly shown that UTDR can indeed be successfully used to monitor membrane fouling in MF, UF and RO, with different membranes, module configurations and foulants.

An unsatisfied market need for a modified plugging index meter, based on ultrasonics, has been determined.

Hence, the following recommendations for further investigations in this area are made:

- The production of a modified plugging index meter based on technology developed in this project should be pursued and a world-patent registered. It should be created at reasonable cost.

- This technology should be applied 'real membrane situations', for example, to improve the production of drinking water that is high in colourants (e.g. natural organic matter).
- Investigations into the use of infrasound for cleaning membranes and for the prevention of fouling of membranes is further strongly recommended.
- The use of ultrasonics as an in-situ, non-invasive monitoring technique, to further investigate a new area of minimising fouling in membranes, i.e. the use of infrasound (low-frequency sound) as opposed to ultrasound (high-frequency sound), warrants consideration.
- The most urgent of these recommendations is what we need to do in the short term, namely the necessary purchase of the latest ultrasonics hardware and then to determine what further improvements can be obtained in terms of resolution, data transforms, speed of data recovery, etc. Specifically, improvements with using LabVIEW technology should be determined.