Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2013.796733

51 (2013) 6401–6406
September

Taylor & Francis
Taylor & Francis Group

Workshop on membrane fouling and monitoring: a summary

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Received 25 February 2013; Accepted 15 April 2013

ABSTRACT

Gathering for a workshop hosted by Robert Field at Oxford University, some of the world's leading experts in membrane fouling presented and discussed recent research progress in this area. The delegates paid particular attention to the key issue of biofouling because of its importance in water treatment and its inherent complexity compared to the mechanics of inorganic particulate fouling. It was remarked that the characterisation and understanding of the extracellular polymeric substances (EPS) in biofilms, and the transparent exopolymers (TEP) involved in their development is still a particularly difficult task, which was a subject present during the entire workshop. Furthermore, advances in characterisation of fouling layers and biofilms utilising ultrasonic time-domain reflectometry (UTDR) and confocal laser scanning microscopy (CLSM) were presented, along with a cautionary perspective on interpretation of data from small sample areas. While membrane fouling reduction is traditionally tackled with a prevention and removal strategy, an alternative "biofilm management" approach was put forward. Although a lot of physical and chemical techniques were presented, delegates also stressed the importance of microbiology for getting the whole picture and the need for a range of carefully selected analytical techniques to do so. This paper summarises the proceedings and discussion at this workshop in September 2012.

Keywords: Membrane fouling; Biofouling; EPS; TEP; Workshop Oxford

1. Introduction

The Presented at the Workshop on Membrane Fouling and Monitoring, 21–22 September, 2012, Balliol College, University of Oxford, UKissue of membrane fouling has drawn the attention of thousands of researchers across the globe. In order to reduce fouling or atleast its negative consequences, it is preferable to first develop a fundamental understanding of the problem. For a number of years, the theory of membrane fouling inferred the build-up of discrete particles and molecules on the surface or within the pore structure of the membrane [1]. Whilst this is by no means invalid, a more complex understanding of fouling is evolving due to biofilms, commonly referred to ad biofouling in which bacteria cell colonies and associated biological material, develop and cover the surface. The adoption of this new concept among the membrane

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Presented at the Workshop on Membrane Fouling and Monitoring, 21–22 September 2012, Balliol College, University of Oxford, UK

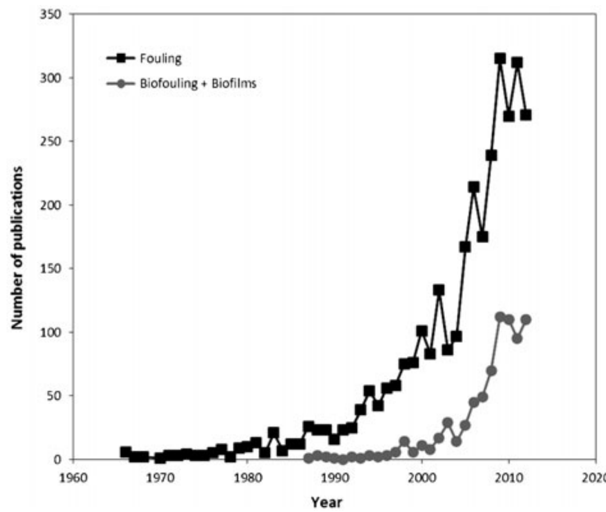

Fig. 1. Publications by year in selected membrane journals (Journal of Membrane Science, Desalination and Desalination and Water Treatment). Search criteria are for appearance in abstract, keywords and titles of "fouling" (squares) and "biofouling" + "biofilm" (circles). Note that some overlap is likely to occur between the two.

technology community appears to have surfaced properly around the mid 1990s, with biofouling research now forming a significant portion of that which focused on membrane fouling. This is demonstrated in Fig. 1, which shows the yearly publications on the subjects fouling and biofouling in this journal, and two other leading journals for membrane studies.

Numerous leading academics in the field of membrane science and technology were invited by Robert Field for a two day workshop in Oxford to discuss current progress on membrane fouling and monitoring. Particular attention was paid towards biofouling and its significance to water treatment. This report presents a summary of the recent work presented by delegates of this workshop.

2. Biofilm development

It is generally accepted now, that biofilms generated in water treatment are made up mostly from a hydrated matrix of extracellular polymeric sub stances (EPS), with less than 5% consisting of actual microorganisms [2]. The EPS also makes the largest contribution to the resistance incurred from biofouling, however the extent to which it does can be dependent on its chemical composition and morphology. As a result of numerous studies of fouling on RO membranes filtering Red Sea water, research led by Jean–Phillipe Croué has suggested, that rather than just becoming more voluminous, biofilms showed increased biodiversity, and protein content for longer filtrations. D.C. Sioutopoulos presented work in which alginate films, analogous to biofilms, exhibited higher resistance when calcium ion concentration was higher.

The biofilm is held to the surface of the membrane by a combination of hydrogen bonding, Van Der Waal's forces, and electrostatic and ionic interactions. The initial adhesion is not necessarily dependent on the flux driven transport of microbes towards the membrane, as indicated by J.S. Vrouwenvelder, who has shown in earlier work that biofilms can proliferate on feed spacers [3] in spiral wound membranes; and reported at this workshop that biofilms were still prevalent on membranes even after experiments without flux. However, Tony Fane showed data which shows where increased flux increased the development of biofilms, with and without spacers, and where spacers reduced the rate of biofouling [4].

The mechanical properties of biofilms were also explored in this workshop. Sioutopoulos presented evidence that alginate films behaved in a more solid-like manner than liquid, a surprising result to some, as the behaviour of biofilms was presumed to be more gel-like. The ionic concentration was also found to have a strong effect on the rheology of these films. Vrouwenvelder has also reported that lower shear rates on the surface where the biofilm grows, causes it to become more "fluffy", making it easier to remove and reducing resistance to tangential flow.

Another major factor in the development of biofilms, and also the subject of a lot of discussion, was the presence of TEPs, whose concentration has been directly linked to the rate of membrane fouling. These gel-like substances are excreted by microbes and self-assembled to form independently suspended particles abundant in almost all aquatic environments [5]. Due to their sticky nature they strongly influence the formation of biofilms, as well as contributing to membrane surface fouling. Tom Berman proposed a "revised paradigm" for the description of biofilm formation, in which TEPs can form a conditioning layer to which cells can adhere before producing EPS and proliferating [6]. The compositional characteristics of TEPs can vary greatly depending on their source, as shown by Harvey Winters, who demonstrated that TEP excreted by marine bacteria (as found in the Red Sea) contained a larger proportion of lower molecular weight glycoproteins than that from phytoplankton. The influence of TEPs on membrane filtration alone was further demonstrated by Zheng and Croué, who investigated the fouling potential of individually isolated biopolymers. The definition of these polymers as effluent organic matter may overlap with that of TEPs to some extent, however the two are currently considered exclusively.

3. Characterisation techniques for membrane fouling

In characterising fouling layers and biofilms are wide variety of techniques which must be combined in order to build a comprehensive, and mechanistic understanding of their growth and removal.

Morphological data in-particular is best inferred with as little disturbance to the system as possible, which has led to a great degree of innovation in the development of non-invasive methods for monitoring fouling [7,8]. In this workshop, Victor Sim presented work performed using ultrasonic time-domain reflectometry (UTDR)–a technique in which reflected ultrasound signals are interpreted to indicate, deposit thickness and density–which has recently been used to demonstrate the glass-transition of gel layers of colloidal silica [9]. Corinne Cabassud has overseen recent work using Laser Sheet at Grazing Incidence [10,11], a form of laser triangulometry which works across the entire width of a flow cell, and explained how results using this and UTDR can differ when measuring the thickness of a flowing cake layer [11]. Vrouwenvelder's work has involved a direct observation method using a "Membrane Fouling Simulator" [12], as well as using nuclear magnetic resonance imaging [13] of spiral-wound membranes. The latter of these has allowed the acquisition of images which demonstrate not only the level of biofouling in specific areas across a large filtration area, but also the local velocities and hence the effect of biofilms on the membrane flux and tangential flow across feed spacers. Vicki Chen also described a direct observation method which has been in use by her group over the last few years to view fouling and removal on single hollow fibres through a microscope [14]. The potential of a commercially available optical device (OPTIQUAD, Krohne Optosens GmbH), which performs spectroscopic measurements at wavelengths from 200 to 4,000 nm, was investigated by H-C Flemming. Although only performing studies of biofilms on non-porous media, he demonstrated that it was capable of making quite reliable measurements of biofilm activity and growth. Another method which could potentially be useful for membrane studies is that used to study TEP and biofilm deposition on surfaces by Berman's research group, in which fluorescence microscopy was used to track TEP aggregation on surfaces in real time [6].

Ex-situ analytical techniques are generally much more established, because they do not require bespoke equipment attached to a filtration rig, their use is much more widespread. Confocal laser scanning microscopy (CLSM) is favoured by all for the identification of specific components and the morphology of fouling layers; however Leiknes gave a cautionary account of the way in which it can provide misleading data. CLSM only provides analysis of a small area ($\sim 0.05\,mm^2$), so multiple samples are required. The severe non-uniformity with which large membrane areas are fouled, especially when dealing with biofouling, means the accuracy of generalised data for an entire system is at the mercy of the sampling process. CLSM and epifluorescence microscopy also rely on correct selection of staining compounds. Electron microscopy and atomic force microscopy (AFM) have been used in morphological characterisation and, through the use of NOM coated probes, Croue's research group is currently studying adhesion strength of foulants on to membranes using AFM. In the first study of its kind, Sioutopoulos presented experiments in which a parallel plate rheometer was used to characterise the complex viscoelastic properties of alginate films on RO membranes.

4. Modelling and simulation

Mathematical modelling is the basis for computational fluid dynamics and molecular simulation which are the two main approaches used to predict fouling, but stochastic approaches [15] and process simulations are also utilised.

Membranes show a different fouling behaviour than plain surfaces due to the permeating substance creating a flow orthogonal to the membrane. Guy Ramon gave an introduction to his refined mathematical method based on the methods developed by Brenner [16] and Goren [17] for calculating particle movement towards a permeable surface [18].

Harry Ridgway presented the "forced molecular dynamics" method which allows for atomistic simulation of the adsorption of biomolecules to the membrane surface. He demonstrated it in his example as an alginate molecule floating over a membrane "runway" where repeatedly occuring small molecular interactions lead to a final attachment of the molecule to the surface. It was acknowledged that this technique, though it is heavily computationally intense, has significant potential for understanding and predicting fouling on membrane surfaces.

Working on modelling of membrane bioreactors (MBR), Parneet Paul presented his recent work on an MBR benchmark model, which would be analogous to the COST benchmark simulation model used for activated sludge wastewater treatment plants.

5. Pre-treatment

Feed water typically contains various concentrations of dissolved matter such as salts and suspended solids, depending on its origin. Pre-treatment is necessary to reduce the foulants and scalants in the feed water, thus improving the efficiency of downstream water treatment operations.

Rodney Komlenic presented a novel pretreatment filter called Disruptor®, a product of Ahlstrom Advanced Filtration. The filter is a non-woven mesh covered with positively charged crystalline boehimite nanofibres, which is able to remove foulants due to electroadsorptive and hydrophobic interaction. These nano-fibres allow the removal of substances smaller than the mesh size.

Reliable evaluation of pretreatment efficiency according to Roger Ben Aim is challenging, but can be achieved by online measurement of assimilable organic carbon (AOC). The AOC content was continuously obtained by a modified rapid bioluminesence measuring method similar to the method used in Berman's group. Using this technique Roger Ben Aim's group compared a granulated active carbon filter (GAC) with an anthracite biofilter, proving the superiority of the GAC filter. Ben Aim highlighted the submerged membrane adsorption bioreactor with powder activated carbon as a pre-treatment unit for reverse osmosis, showing that it has very low fouling potential while producing suitable feed water quality at a decent flux.

6. Fouling monitoring and control

Moving towards a better fouling control in an active plant brings up the necessity for monitoring solutions. Sim and Fane suggested that the UTDR method they presented may be used as an early warning system for membrane fouling. They showed promising results that indicated the presence of fouling before identified by a rise in TMP. Fane also expressed that evaluation experiments of an early warning device (built by Inphaze Ltd. [19]) using Electrical Impedance Spectroscopy were also underway.

Robert Field provided some viewpoints on integrity sensors, early warning devices used to detect perforations in membranes; for example when permeate contamination might occur when 1 of 1,500 hollow fibres breaks during a filtration. In recent work on such devices Tony Fane's group developed a technique in which small filtration cells are placed on side-streams taken from permeate lines of a filtration system [20,21]. Field presented theory in which this could be best achieved, proposing three different filtration conformations: a simple dead end filtration, a two stage series of membranes, and a two-stage membrane and valve configuration. A performance metric C, the ratio of the difference in sequential pressure drops to the overall pressure drop across the cell as proposed by Krantz [21], was presented as a means of indicating whether fouling characteristic of permeate contamination has occurred. Some open questions remained from this on which type of fouling was desirable (with respect to the sequential blocking laws), and whether excellent capture could be achieved using cross-flow cells. Microsieves were also proposed as a possible alternative to membranes in such devices.

7. Cleaning strategies

The attitude towards the cleaning of biofouling is one of fatalism amongst microbiologists. Hans–Curt Flemming proposed that in water treatment, biofilms cannot be avoided and that an attitudinal shift from kill-all disinfection towards a more pragmatic approach of biofilm "management" is necessary. Disinfection does not necessarily remove a biofilm, as the dead cells are still held on the surface by the EPS. Whilst mechanical removal is still thought of as the best removal method, enzymatic cleaning holds some potential, although will only work to break apart very specific components within the film. Backwashing studies performed by Vicki Chen's group also showed how a more cunning approach towards foulant removal may be required. She noted that while optimising the frequency and duration of backwash cycles helped to remove alginate biofilms from UF membranes, the most effective removal of alginate was achieved using DI water as the backwash water. It was pointed out however, that on an industrial scale this would be neither practical nor economically viable. Vrouwenvelder presented evidence that biofilms on feed spacers where harder to remove mechanically, and posed a greater resistance to flow when formed at higher cross-flow velocities. He proposed that the overall filtration performance of spiral wound membrane systems susceptible to biofouling could be improved by operating at lower cross-flow velocities to reduce feed side pressure drop towards the inlet side of the module. Sher Jamal Khan also proposed innovations in cleaning techniques, presenting a study in which a membrane-based septic tank was tested. The cleaning protocol used involved sun drying, followed by brushing, washing and tap water filtration; and Khan reported sustained operation over multiple fouling/cleaning cycles up to 11 LMH. The obvious drawback is that this methodology is only proven for hot climates.

8. Summary

The final presentation of the workshop was delivered by Graeme K. Pearce, of Membrane Consultancy Associates Ltd. who applied a cost-modelling approach to determine the viability of ceramic membranes in place of polymerics. Since the 1990s, popularity of ceramics over polymerics has decreased due to their higher capital costs, however their higher flux and durability coupled with a lower fouling potential, may make them a more competitive process than originally thought. The cost-model took into account many aspects of a membrane process including the price and lifetime of membranes, the water, chemicals and cleaning costs, flux and power requirements, and the auxiliary components associated with each. Pearce showed that polymeric membranes systems are still likely to work out cheaper overall, and that ceramics would have to confer a further 10% increase in flux to the overall process before they become more economically viable than polymerics. It was conceded however that long-term operational data for ceramic membranes is scarce, despite a 20 year lifespan being estimated for ceramic modules.

A short discussion at the end of the workshop reflected this pragmatic philosophy towards mem brane process optimisation. A number of issues and questions were put forward, looking towards the future of fouling remediation:

With regards to the numerous advanced analytical techniques available, including *in-situ* and *ex-situ* morphological and chemical characterisations, an important question is of how to select the most relevant methods for a particular application. The impact of such a decision filters down to issues which are perhaps unique to specific filtrations and even particular plants, wherein op timum pretreatment and cleaning protocols could vary greatly.

The new approach of management rather than eradication of biofilms suggested by Hans-Curt Flemming was reiterated, suggesting that their resistance could be influenced. A better understanding of how EPSs cause flux resistance is required, with emphasis on the role of cake and biofilm enhanced osmotic pressure (CEOP and BEOP). This again highlights the need for a well-informed selection of analytical techniques.

Another key issue raised was that there is little communication of the needs of industries which could help to influence the direction of current research. One perspective offered from the industrial camp was that issues with membrane materials remained largely unresolved, in that modified hydrophilised and low-energy membranes did not perform well practically.

A. Karabelas proposed a global simulator for RO plants, based upon shared data from plants around the world. It was acknowledged that this would be a very ambitious pursuit, requiring reverse osmosis system analysis data for individual modules of RO systems. Research goals in this field have shifted from the eradication and prevention of fouling, towards management and understanding. This new approach may help drive cleaning “from art to science” and pave the way towards less energy intensive, but more effective cleaning protocols.

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