Powdered activated carbon and biofiltration improve MF performance: Part II

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This article shows how the use of high-dose powdered activated carbon and biofiltration are able to improve the performance of membrane-based microfiltration systems. The first part of this feature, which was published in the May 2007 issue of *Membrane Technology*, provides an overview of the study, materials and methods, and experimental design and operational conditions of the reactors. The second instalment, which appears here, discusses the results of this study.

Three microfiltration (MF) membrane systems were operated for approximately 33 days with periodic back-washing for fouling control.

Reactor 1 was a control, reactors 2 and 3 contained 40 g/l of powdered activated carbon (PAC), and reactor 3 also was fed only biofilter treated water. No modifications were made to the method of operation or the feed water during these periods.

Transmembrane pressure of membrane modules

Operating at a constant flux, all three reactors accumulated transmembrane pressure (TMP) over the course of the study.

The TMP records of the membrane modules are shown in Figure 1. As can be seen in this figure, the system treating raw water without the benefit of PAC accumulated TMP more rapidly than the other two systems. Once the TMP of membrane inside reactor 1 (raw and no PAC) reached more than 50 kPa (0.5 bar), all reactors were stopped. However, the TMPs of other reactors (reactor 2 and 3) dosed with PAC, could have been operated for a longer period.

Cake and gel layer formation

Prior work[1] with PAC and MF showed that cake and gel layer formation were the primary mechanisms of membrane fouling. The adsorbed and attached materials on the PAC were responsible for the loss in performance.

During this experiment, a thick cake layer was not found on the outer surface of any fouled membrane. During physical cleaning, cake was observed to accumulate primarily between membrane fibres, and at the ends of the fibres, where aeration could not reach. It is likely that aeration contributed significantly to the removal of cake fouling in this system.

Role of carbohydrate on membrane performance

Extracellular polymeric substances (EPS) inside the membrane biofoulant possess many important functions, including anchoring the micro-organisms near food sources, protecting them from dehydration and toxic substances, and providing ion exchange properties because of the negatively charged surface functional groups, which allow them to bind cationic species such as heavy metals.[2]

Lee *et al.* (2001)[3] reported that the cake layer could consist of a variety of components – micro-organisms and various inorganic and organic substances including EPS. The slow decline phase of membrane flux results from establishment of an

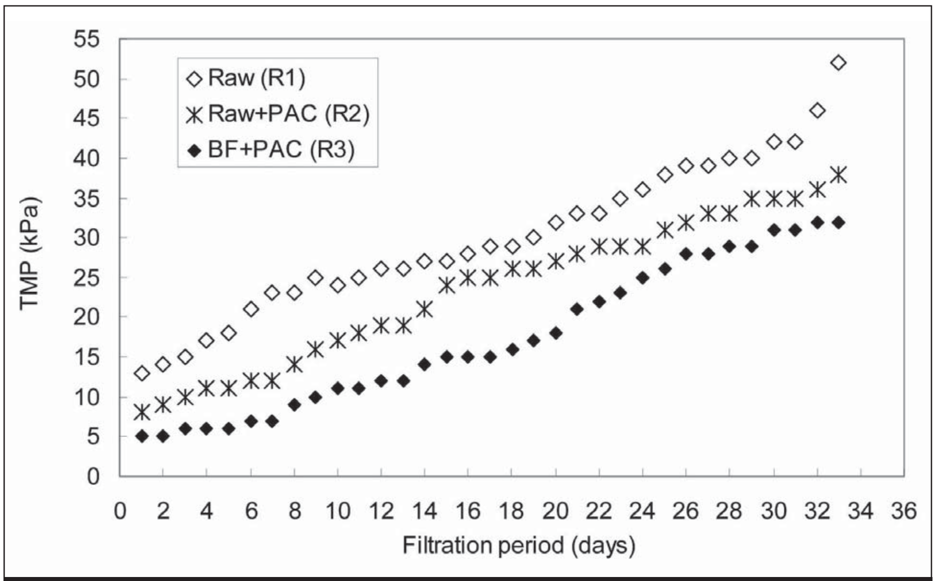

Figure 1. Transmembrane pressure (TMP) records of membrane modules inside the reactors.

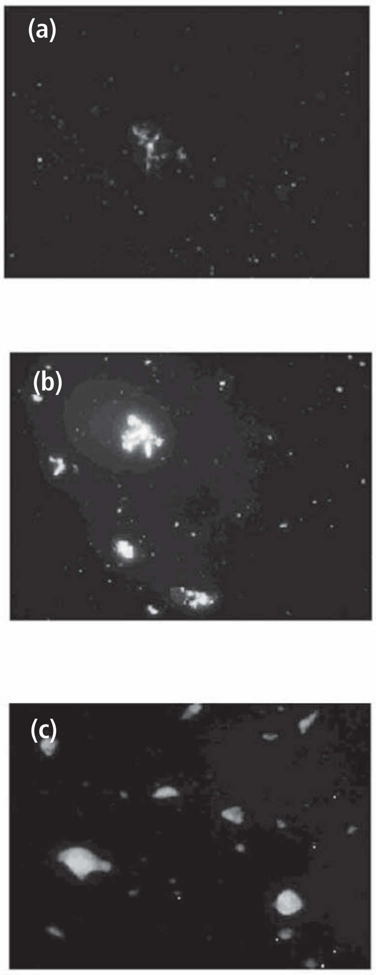

Figure 2. Extracted extracellular polymeric substances (EPS) from samples (*a*) inside R1, (*b*) inside R2 and (c) inside R3 were labelled by jack bean lectin (D(+) glucose and D(+) mannose).

Lectin	Observed day	Average mean value of light intensity ± standard error of mean		
		Reactor 1	**Reactor 2**	**Reactor 3**
Red kidney bean	Day-1	2.01 ± 0.09	1.23 ± 0.08	0.56 ± 0.04
	Day-18	3.21 ± 0.06	1.89 ± 0.07	0.89 ± 0.04
	Day-33	4.11 ± 0.05	2.11 ± 0.06	1.24 ± 0.03
Peanut	Day-1	1.01 ± 0.06	0.87 ± 0.03	0.42 ± 0.02
	Day-18	1.65 ± 0.04	1.01 ± 0.02	0.69 ± 0.04
	Day-33	2.13 ± 0.02	1.43 ± 0.01	0.99 ± 0.03
Gorse/Furze	Day-1	2.41 ± 0.06	1.54 ± 0.04	1.11 ± 0.06
	Day-18	4.21 ± 0.05	2.01 ± 0.02	1.87 ± 0.05
	Day-33	5.36 ± 0.05	2.87 ± 0.07	2.68 ± 0.03
Coral tree	Day-1	1.89 ± 0.05	0.99 ± 0.03	1.21 ± 0.02
	Day-18	3.24 ± 0.02	1.12 ± 0.02	0.97 ± 0.05
	Day-33	5.34 ± 0.06	2.34 ± 0.04	1.56 ± 0.06

Table 1. The average mean values of light intensity of red kidney bean, peanut, gorse/furze and coral tree for samples inside reactors. Standard error of mean values are also shown.

Lectin	Average mean value of light intensity ± standard error of mean			
	Reactor 1	**Reactor 2**	**Reactor 3**	**Activated sludge**
Red kidney bean	47.23 ± 0.55	45.12 ± 0.42	31.12 ± 0.48	47.86 ± 0.65
Peanut	30.12 ± 0.09	26.34 ± 0.72	36.23 ± 0.59	18.72 ± 0.58
Gorse/Furze	25.34 ± 0.37	24.35 ± 0.48	18.56 ± 0.77	38.12 ± 0.41
Coral tree	47.23 ± 0.59	49.23 ± 0.82	54.36 ± 0.68	35.13 ± 0.87

Table 2. The average mean values of light intensity of red kidney bean, peanut, gorse/furze and coral tree for the foulants on membranes at Day-34 and activated sludge. Standard error of mean values are also shown.

equilibrium condition in which biofilm growth and EPS production are balanced by biofilm loss (cell detachment or sloughing) caused by hydrodynamic shear at the solution-biofilm interface.[4]

Figure 2 shows the images of extracted EPS from samples inside reactor 1, 2 and 3 after labelling by jack bean lectin (D(+) glucose and D(+) mannose). For the other lectins, similar images were observed during investigation and their corresponding mean values of light intensities were calculated.

Because each lectin has different light scattering and absorption properties, it is not possible to compare intensities among lectins in a manner that quantifies the carbohydrate content.

Comparisons were performed only among samples labelled with the same lectin. **Figure 3** illustrates the mean value of light intensity of jack bean lectin for samples inside the three reactors, for the foulants on the membranes, and in the activated sludge as a control. The average mean values of light intensities of other lectins for samples inside the three reactors and for the foulants on the membranes are summarized in **Table 1** and **Table 2**, respectively.

For all of the fluid samples and for all five lectins, the highest light intensities were observed for the sample inside reactor 1 and the lowest intensities were found inside reactor 3.

The light intensities of lectins, except peanut (D(+) galactose) and coral tree (N-acetyl-D-galactosamine and D-galactose), were the highest in samples from the membrane foulant in reactor 1, which were consistent with the bulk solution data.

Once the specific carbohydrate was adsorbed on PAC (because of the interaction with micro-environment[5] on PAC particle) it was degraded or changed into different components. Biofiltration of surface water played significant role in removing these carbohydrates. Pretreatment of the feed water can be a useful strategy for reduction or mitigation of these fouling effects.[6]

For the peanut and coral tree lectins, the highest concentration of lectins was found in the membrane foulant from Reactor 3. The intensity of all carbohydrates is 40–60 times higher in the foulants than the samples in the bulk phase, which supports the concept that accumulation of carbohydrates on the membrane surface contributed to flux decline.

However, TMP records (Figure 1) indicate that the addition of PAC and biofiltration decreased the resistance to filtration. Thus, the presence of the carbohydrate corresponding to coral tree (N-acetyl-D-galactosamine and D-galactose) did not appear to correlate with increased membrane resistance.

The long-chain carbohydrates act as bonding agents among the particles and other foreign dissolved and suspended matter, which create a stronger bridge on the available surface of the membrane and also the particles, resulting in increased TMP.[4] PAC could make enough porous spaces in front of the membrane to hold a very high quantity of carbohydrates.

Impact of protein on membrane performance

The data on proteins in the system are somewhat in contrast to those covering carbohydrates.

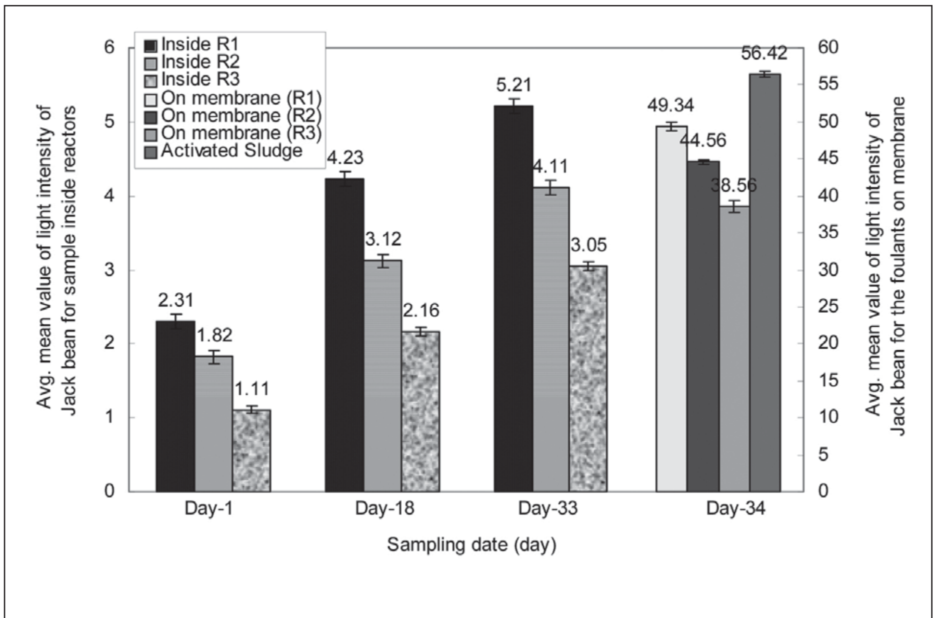

Figure 3. Average mean value of light intensity of jack bean lectin (D(+) glucose and D(+) mannose) for sample inside reactors and for the foulants on the membrane. The error bars indicate the standard error of mean for each sample.

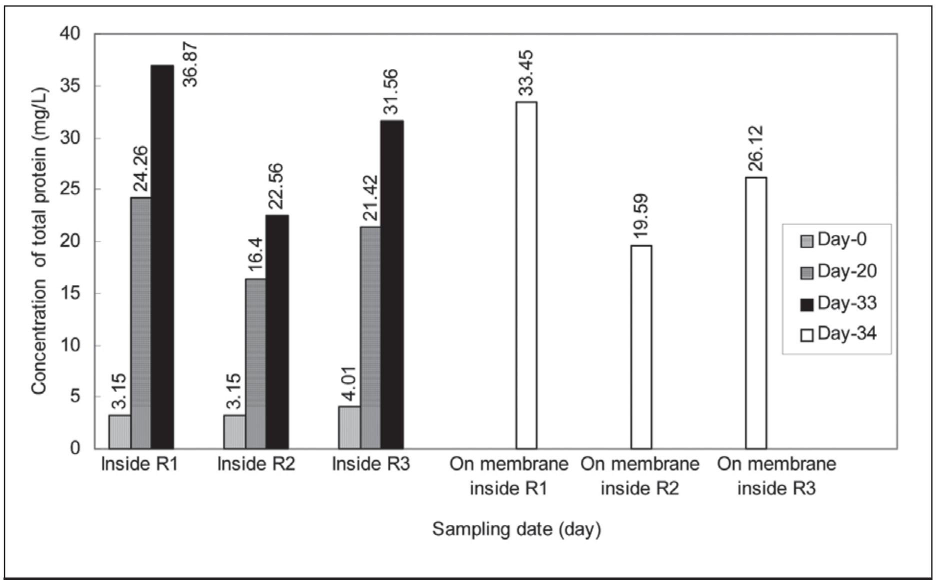

Figure 4. The concentration of total protein extracted from the samples inside the reactors and also from the foulants on the membranes.

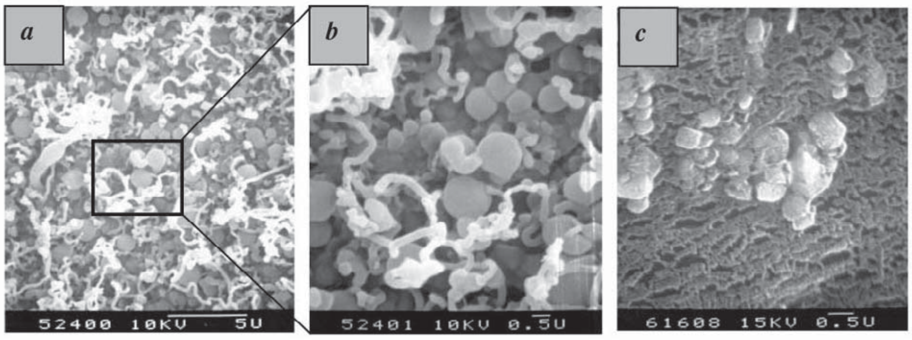

Figure 5. Scanning electron microscopy images of (*a*) extracellular polymeric substances on the fouled membrane inside reactor 3 after physical cleaning; (*b*) a close-up image of (*a*); and (*c*) the membrane surface after chemical cleaning with powdered activated carbon (PAC) particles blocking some pore spaces of the membrane.

By comparing the protein concentrations of the sample inside the reactors on Day-0, Day-20 and Day-33 (**Figure 4**), protein in the bulk fluid accumulated relatively constantly – initially starting at a low value.

The carbohydrate accumulation was much less linear (Figure 3), being roughly twice as high after 18 days than after only 1 day of operation. The protein concentrations from biofoulant samples are of the same order of magnitude as the bulk fluid samples, while the carbohydrate intensities on the membranes is a full order of magnitude higher than in the bulk fluid.

The biofoulants were extracted from the membrane surfaces, but 100% recovery was impossible. So the real amounts of protein inside membrane foulants were higher than reported in Figure 4. The influents were carrying less protein, but inside the reactors and in the foulants of all membrane modules, the total amount of protein was 5–10 times more, which indicates that because of the microbial degradation and consecutive accumulation of proteins from the source waters, there were heavy loads of protein on the membranes.

More protein accumulated on the membrane and in the bulk solution in reactor 3 than in reactor 2, but this is consistent with a higher protein concentration in the feed to reactor 3.

Biofiltration could reduce suspended solids by more than 80%, but it also produced more proteins because of the microbial degradation inside the bioreactor. Thus, it appears that the proteins in this system (reactor 3) were relatively stable (not significantly degraded), but also were not preferentially adsorbed on the membrane surface because of the presence of PAC. Conversely, the carbohydrates showed a higher affinity for the membrane surface, but were also apparently more labile than the proteins.

Scanning electron microscopy images

Figure 5 shows scanning electron microscopy (SEM) images of a fouled membrane inside reactor 3, after physical and chemical cleaning.

Physical cleaning by Milli-Q water could not remove a significant part of extracellular materials (Figures 5*a* and 5*b*), but chemical cleaning could clean them completely. However, PAC particles blocked the pores of membrane in a scattered manner (Figure 5*c*), which means that neither back-washing nor chemical cleaning could completely recover the pore spaces of membrane.

Compared with other SEM images of membranes inside reactor 1 and reactor 2 (data not shown), it was found that the addition of PAC and the use of water treated by biofiltration gave a higher proportion of unblocked pore spaces.

Conclusion

The use of a high dose of PAC inside a MF unit resulted in improved performance and lower carbohydrate and protein concentration on the membrane and in the bulk fluid.

Biofiltration prior to treatment further improved membrane performance and reduced all bulk solution carbohydrates, but produced higher concentrations of protein and some carbohydrates on the membrane surface. Thus, increased concentrations of some compounds correlated with increased physical fouling (as measured by TMP), while other compounds do not appear to affect fouling.

Overall, however, the use of high-dose PAC and biofiltration of feedwater show promise for improved performance of membrane systems.

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(The full title of this paper, as submitted by the authors is: 'Effect of carbohydrates and protein on the biofouling formation of microfiltration membrane combined with a high-dose PAC.')

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Research Trends

Reverse osmosis system design

The design of various multi-stage reverse osmosis (RO) systems under different feed concentration and product specifications is covered by this work. An optimization method using the process synthesis approach to design an RO system has been developed. First, a simplified superstructure that contains all the feasible design elements used in a current desalination process is presented. It offers extensive flexibility towards optimizing various types of RO systems and thus may be used for the selection of the optimal structural and operating schemes. A pressure vessel model that takes into account the pressure drop and concentration changes in the membrane channel is also given to simulate multi-element performance in the pressure vessel. Then the cost-equation, relating the capital and operating cost to the design variables, as well as the structural variables of the designed system, are introduced in the objective function. Finally, the optimum design problem can be formulated as a mixed-integer non-linear programming (MINLP) problem, which minimizes the total annualized cost. The solution to the problem includes optimal arrangement of the RO modules, pumps, energy recovery devices, the optimal operating conditions and the optimal selection of types and number of membrane elements. The effectiveness of this design methodology has been demonstrated by solving several sea-water desalination cases. Some of the trends of the optimum RO system design are also presented.

Y.-Y. Lu, Y.-D. Hu, X.-L. Zhang, L.-Y. Wu and Q.-Z. Liu: *J. Membrane Science* **287**(2) 219–229 (15 January 2007).
DOI: 10.1016/j.memsci.2006.10.037

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Cross-linked lyotropic liquid crystal membranes

In this study, novel, nanostructured lyotropic liquid crystal polymer membranes were fabricated, characterized and tested for their performance in separation processes involving CO_2, N_2, O_2, CH_2 and H_2. Freestanding, cross-linked films with and without an ordered hexagonal nanostructure, but with the same chemical composition, exhibit different gas solubility, diffusion and permeability properties. The presence of an ordered nanostructure was observed to slow diffusion of all gases, but increase solubility and permselectivity in CO_2-based separations. In systems with an ordered nanostructure, CO_2 permeability was 5.2 barrers and CO_2/N_2 separation selectivity was 27. For membranes with the same chemical composition, but lacking an ordered hexagonal nanostructure, permeability rose slightly to 6.9 barrers, but CO_2/N_2 separation selectivity dropped to 21. A similar trend was also observed for CO_2/CH_4 separation. In separation processes with gases other than CO_2, no permselectivity differences were observed between the two membranes. A model explaining these trends is also presented.

J.E. Bara, A.K. Kaminski, R.D. Noble and D.L. Gin: *J. Membrane Science* **288**(1–2) 13–19 (1 February 2007).
DOI: 10.1016/j.memsci.2006.09.023

Modelling submerged hollow-fibre membrane filtration

A model for submerged 'outside–in' hollow-fibre microfiltration/ultrafiltration (MF/UF) for wastewater treatment forms the subject of this paper. The model aims to integrate the major factors influencing medium-scale granularity. In particular, the model covers the geometry of the system, the hydrodynamics of the feed and of the permeate flow, and the filtration resistance. The filtration resistance model considers membrane resistance, pore blocking, formation of the cake layer, poly-dispersed particles, biofilm formation and concentration polarization. The model is thoroughly analyzed in simulation and sensitivity studies. The role of uncertain parameters is discussed. The influence of important operational parameters of the biological system and of the membrane aeration rate on the filtration performance is also investigated. The model provides insight into the MF/UF filtration process in membrane bioreactors and the interplay of the related physical, chemical and biological phenomena.

J. Busch, A. Cruse and W. Marquardt: *J. Membrane Science* **288**(1–2) 94–111 (1 February 2007).
DOI: 10.1016/j.memsci.2006.11.008

Novel membrane reactor for ozone water treatment

A novel membrane reactor was designed for the ozonolysis of refractory organic pollutants in water.