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Development of a setup to enable stable and accurate flow conditions for membrane biofouling studies

Szilárd S. Bucs^{a,*}, Nadia Farhat^a, Amber Siddiqui^a, Rodrigo Valladares Linares^a, Andrea Radu^{b,c,1}, Joop C. Kruithof^c, Johannes S. Vrouwenvelder^{a,b,c,*}

^aWater Desalination and Reuse Center, Division of Biological and Environmental Science and Engineering, King Abdullah University of Science and Technology, Thuwal 23955-6900, Saudi Arabia, emails: szilard.bucs@kaust.edu.sa (S.S. Bucs), nadia.farhat@kaust.edu.sa (N. Farhat), amber.siddiqui@kaust.edu.sa (A. Siddiqui), rodrigo.valladares@kaust.edu.sa (R. Valladares Linares), johannes.vrouwenvelder@kaust.edu.sa (J.S. Vrouwenvelder)

^bFaculty of Applied Sciences, Department of Biotechnology, Delft University of Technology, Julianalaan 67, 2628BC, Delft, The Netherlands, emails: andrea.radu@empa.ch (A. Radu), j.s.vrouwenvelder@tudelft.nl (J.S. Vrouwenvelder) ^cWetsus, European Centre of Excellence for Sustainable Water Technology, Oostergoweg 9, 8911 MA Leeuwarden, The Netherlands, emails: joop.kruithof@wetsus.nl (J.C. Kruithof), hans.vrouwenvelder@wetsus.nl (J.S. Vrouwenvelder)

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ABSTRACT

Systematic laboratory studies on membrane biofouling require experimental conditions that are well defined and representative for practice. Hydrodynamics and flow rate variations affect biofilm formation, morphology, and detachment and impacts on membrane performance parameters such as feed channel pressure drop. There is a suite of available monitors to study biofouling, but systems to operate monitors have not been well designed to achieve an accurate, constant water flow required for a reliable determination of biomass accumulation and feed channel pressure drop increase. Studies were done with membrane fouling simulators operated in parallel with manual and automated flow control, with and without dosage of a biodegradable substrate to the feedwater to enhance biofouling rate. High flow rate variations were observed for the manual water flow system (up to $\approx 9\%$) compared to the automatic flow control system (<1%). The flow rate variation in the manual system was strongly increased by biofilm accumulation, while the automatic system maintained an accurate and constant water flow in the monitor. The flow rate influences the biofilm accumulation and the impact of accumulated biofilm on membrane performance. The effect of the same amount of accumulated biomass on the pressure drop increase was related to the linear flow velocity. Stable and accurate feedwater flow rates are essential for biofouling studies in well-defined conditions in membrane systems.

Keywords: Feed flow rate; Biofouling; Pressure drop; MFS operation system; Controlled biofouling studies

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^{*}Corresponding authors.

¹Chair of Building Physics, Swiss Federal Institute of Technology Zürich (ETHZ), Stefano-Franscini-Platz 5, 8093 Zürich, Switzerland.

1. Introduction

Biofouling, the unwanted deposition and growth of biofilms [1], is a problem encountered in most membrane installations in the water industry [2]. Extensive pretreatment and/or chemical dosage to feedwater can prevent particulate fouling or scaling, however, biofouling remains a major problem, especially in spiral wound membrane systems [3–9].

Biofilm growth in membrane systems and the impact on membrane performance is affected by several interrelated factors, such as the availability of biodegradable nutrients, hydrodynamics [10–13], feed spacer design, and surface material [14], and it is not directly related to parameters measured in the feedwater such as total organic carbon (TOC) concentration and bacterial cell number [8]. Therefore, there is a need for systematic, controlled laboratory studies with a well-designed membrane biofouling monitor system, representative for full-scale applications.

Many monitors have been developed to study biofilm growth [15–19]. Most monitors were designed to simulate biofouling in heat exchangers, cooling towers, and water distribution networks [20,21], and were not representative for biofouling in membrane systems. Although the mentioned applications share the same problem, biofilm development and the impact on performance might be entirely different. For example, a biofilm monitor developed to study the biofouling growth in a water distribution network will not be for biofouling development representative in membrane filtration installations.

Fouling in membrane-based water treatment installations causes an increase in normalized pressure drop over the feed channel and/or reduction in normalized flux and/or increase in salt passage [22]. In order to enable improved plant performance, monitor studies should be carried out to get a better understanding of the biofilm development process, in order to formulate more suitable control strategies.

A membrane biofouling monitor system is defined as a device, where water-containing nutrients passes through a flow cell, containing a membrane and a feed spacer to which micro-organisms attach and grow to form a biofilm. Operational data can be obtained from the monitor like pressure drop increase, water flux, and feedwater flow rate, and allows visual observation of fouling development. For a representative application, the monitor should resemble the spiral wound membrane element, in terms of materials and hydrodynamic conditions: the flow distribution should be uniform, and the pressure drop should be similar to spiral wound elements and should have the same feedwater composition. The relationship between linear flow velocity and pressure drop for the monitor should be representative of the spiral wound membrane element.

Currently, containing monitors membranes and feed spacers, differing in terms of size and operation mode are available to study the biofilm development in spiral wound membrane systems [23-25]. Many of these monitors have shown to be suitable for biofouling studies in membrane filtration systems, but require close attention during operation [26,27]. The driving force in applied membrane fouling simulator (MFS) setups to generate flow through the MFS is the drinking water distribution network pressure. The flow rate is controlled manually by a valve installed at the outflow side of the flow cell. During the experiments, the flow rate is measured manually, and the valve is adjusted to keep a constant flow rate. When fouling occurs, flow adjustments have to be more frequently done, making these monitor setups difficult to operate.

Therefore, there is a need for a system to enable constant flow operation and accurate monitoring of operational parameters of small-scale membrane monitors. Such a system should provide a constant feed flow under biofilm accumulation conditions in the monitor and accurate measure of the pressure drop across the flow channel (Table 1).

The objective of this study was to develop a system to provide an accurately measured, stable water flow through the MFSs, enabling to study biofouling in spiral wound membrane systems systematically, under well-controlled and defined conditions.

2. Materials and methods

2.1. Description of the developed biofilm monitor setup

The biofilm monitor setup was equipped with a water pump, pressure dampener, thermometer, flow meter, flow cell, differential pressure transmitter, and a back pressure valve to enable unit pressurization (Fig. 1). All sensors and the feedwater pump were connected to a central computer (TAC Vista Server). The server logged all data received from the sensors and also regulated the feedwater transport pump.

The water flow through the flow cell was provided by a pump (metering diaphragm pump, Hydra-Cell G20, Wanner Engineering, USA). To eliminate pulsation in the water flow, a pressure dampener was installed right at the outlet of the pump. The pump can be connected directly to a water distribution network or a break (equalization) tank. According to the manufacturer's specifications, the maximum water flow rate of the pump is 230 L h⁻¹ at a pressure of 17 bar. Table 1

Inventory of the required features, current status, and the developed system for operation of membrane fouling simulators (MFSs)

	Required features of the system	Current status	Developed system
1.	Stable, constant water flow	Manual reading and frequent manual adjustments	Automated system with flow sensor and continuous automatic adjustments
2.	Adjustable feed pressure for permeate production	Depending on the feedwater pressure (1–4 bar)	The system can be operated between 0 and 17 bar
3.	Accurate measurements and readings of operation parameters	Manual reading with high time frames	Automated adjustable reading frequency from 1 s up to 24 h
4.	Easy and flexible operation	Need for feed pressure and operation parameter readings	Operates without feed pressure and can be manipulated remotely
5.	Compatibility with current flow cells to study fouling in spiral wound systems	Compatible only with low pressure cell, without permeate production	Compatible with current flow cells



Fig. 1. Schematic representation of the system for MFS operation: (a) with manual flow control and the (b) developed setup with automatic flow control and data logging system.

A paddle wheel flow sensor (Flow X3, Italy) was installed on the outlet tube of the pump after the pressure dampener. The flow sensor was connected to the server, and the flow was monitored continuously. A preset flow rate was maintained by increasing or slowing down the rotation speed of the water pump according to the flow sensor signal by the server. Thus, a constant flow rate can be maintained automatically, even when fouling causes a pressure drop increase in the flow cell.

Dosage of chemicals was provided by a diaphragm metering pump (Stepdos 08, KNF Lab, Germany) which can operate at low flow rates $(0.08-80 \text{ mL min}^{-1})$.

A ceramic differential pressure sensor (Endress + Hauser PMD70, Germany) was connected to the flow

12895

cell inlet and outlet. The pressure drop over the flow cell was measured hourly and logged on the server.

The unit can be operated with different flow cells up to a pressure of 17 bar, allowing the study of biofilm formation in filtration systems at different pressures, flow rates, and with or without permeate production.

On the flow cell outlet, a back pressure valve was installed to control the working pressure of the MFS unit.

2.2. System validation

The MFS [24] was used in this developed setup to validate the system. Commercially available reverse osmosis (RO) membrane sheets and 31 mil (787 μ m) thick feed spacers were used. To create representative hydrodynamics in the flow cell, the flow rate was set at 16 L h⁻¹, resulting in a linear flow velocity of 0.163 m s⁻¹. Tap water was used as the feed for the units. A solution of sodium acetate, sodium nitrate, and sodium dihydrogen orthophosphate in a mass ratio C:N:P of 100:20:10, respectively, was employed with a C concentration in the feedwater of 400 μ g/L.

2.3. Evaluation of the fouling material

After eight days of operation and a significant pressure drop increase over the unit with substrate dosage, the flow cells were opened, and the membrane and feed spacer were analyzed. Adenosine triphosphate (ATP) and TOC were measured to quantify the accumulated biomass. Energy-dispersive X-ray (EDX) measurements were carried out to determine inorganic deposition and screen for biofilm accumulation on the membrane surface.

3. Results

A system was developed to provide an accurate and stable water flow to MFSs. Comparison studies

were performed with the *manual* flow control system (used in earlier MFS studies) and the developed *automatic* flow control system to assess (i) feedwater flow rate stability, (ii) feed channel pressure drop, and (iii) biofilm accumulation. Studies were done with and without dosage of biodegradable organic nutrients to the MFS feedwater to enhance biofilm formation. A summary of the studies is shown in Table 2.

3.1. Evaluation of manually controlled reference system

Earlier investigations with the manually controlled flow system have shown varying water flow rates in time passing through the monitors, especially, when significant biofilm accumulation on the feed spacer and membrane sheets in the monitor occurred. Delayed correction of the flow rate affected the pressure drop over the monitor. To quantify the variation of flow rate and its impact on pressure drop development, parallel studies were done using (i) a monitor without nutrient dosage and (ii) a monitor with nutrient dosage. The water flow rate and pressure drop were monitored in time.

The flow rate for the monitor without nutrient dosage showed variations up to $\pm 3\%$ of the set feed flow rate (16 L h⁻¹ and 0.16 m s⁻¹) during the research period (Fig. 2(a)), while the pressure drop over the monitor was relatively constant with time (Fig. 2(b)).

For the MFS with nutrient dosage, during the first four days of the study, the flow rate showed variations up to $\pm 3\%$ and a relative constant pressure drop. After four days, higher flow rate variations up to $\pm 9\%$ (Fig. 2(a)) were found corresponding to stronger pressure drop variations (Fig. 2(b)). In other words, the manual flow control system was influenced by the impact of biofilm formation, resulting in significant variations in terms of flow rate and corresponding pressure drop (up to $\pm 9\%$). Systematic biofouling studies under well-controlled conditions require a

Table 2

Experimental conditions for the studies to evaluate the suitability of the developed system for biofouling studies with constant water flow

Description	Aspect studied	Nutrient dosage	Results
Manual flow control reference system	Flow accuracy, stability, and pressure drop	With and without	Fig. 2
Automatic flow control system (developed)	Flow rate accuracy Flow accuracy, stability, and pressure drop	Without With and without	Fig. 3 Fig. 4
Comparison of manual and automatic flow control in biofilm formation studies	Pressure drop and biofilm accumulation	With and without	Fig. 5



Fig. 2. Feedwater flow rate (a) and pressure drop in time (b) over the MFS systems with manual flow control and fed with water without (MS-) and with supplemented (MS) biodegradable substrate.

more accurate and stable water flow. Therefore, an automatic flow control system was developed.

3.2. Evaluation of the automatic flow control system

The developed automatic system for feedwater flow control was evaluated on the (i) accuracy of the flow rate (3.2.1), (ii) flow stability, and (iii) pressure drop development with and without nutrient dosage (3.2.2).

3.2.1. Flow rate accuracy

The accuracy of the automatic water flow control system was determined by comparison of set point and gravimetrically measured flow rates for the range $10-20 \text{ L} \text{ h}^{-1}$ (equal to a linear velocity of $0.10-0.20 \text{ m s}^{-1}$). Gravimetric flow measurements were carried out to validate the accuracy of the automatic system. The set point and gravimetrically measured flow rates showed a linear relationship for the range of $10-18 \text{ L} \text{ h}^{-1}$ (Fig. 3). Less than 1% flow deviation was observed. A good correlation was found between the automatic system set flow and the gravimetrically measured flow rates. The automatic system was shown to be suitable to provide and maintain an accurate flow rate without biofilm accumulation.



Fig. 3. Correlation between preset and measured flow rate of the developed automated system for the range $10-20 \text{ L h}^{-1}$.

3.2.2. Flow stability during biofilm development

The automatic system for MFS operation was evaluated by operating monitors in parallel (i) without and (ii) with nutrient dosage to the monitor feedwater. The flow rate and pressure drop were monitored in time.

Irrespective of nutrient dosage to the automatic system, flow rate variations smaller than 1% were observed during the whole research period (Fig. 4(a)). In time, (i) without nutrient dosage the pressure drop over the monitor was constant (Fig. 4(b)), while (ii) with nutrient dosage the pressure drop increased sharply. During the research period, the automatic system enabled an accurate, stable water flow with variations smaller than 1% of the set flow rate.

3.3. Comparison manual and automatic flow control in biofilm formation studies

To determine the influence of both manual and automatic flow regulation on pressure drop development and biofilm accumulation, monitor studies were performed. Two manually regulated monitors, and two automatically regulated monitors were run in parallel with and without nutrient dosage.

With nutrient dosage, the pressure drop increased for both the manual and automatic system (Fig. 5(a)). At the end of the eight day research period, the pressure drop increase was lower for the manual system compared to the automatic system (Fig. 5(b)), while the same amount of accumulated biomass (ATP and TOC) was found for both the systems (Fig. 5(c) and (d)). SEM–EDX measurements of the feed spacer and membrane surface from the monitors showed the (i) absence of inorganic deposition in all systems and (ii) presence of biofouling in the systems fed with nutrient dosage.

The flow rate variations of the manual system caused a significantly lower pressure drop than the automatic system with the more accurate flow control. The automatic flow control system provided an accurate and constant feed flow rate to the flow cell irrespective of fouling accumulation.

4. Discussion

The main objective of this study was to develop a system which can provide a constant water flow to the MFSs, enabling biofouling studies in MFS units under well-defined conditions. The developed automatic system was compared with a manually flow controlled system used in previous studies [11,28–30]. For these two systems, water flow rate variations, and its impact on pressure drop increase with and without biomass accumulation were compared (Figs. 2, 4 and 5). Much lower flow variations were observed for the automatic water flow system (<1%) compared to the manual system (up to $\approx 9\%$). It was shown that fluctuations in feedwater flow rate directly impacts the measured performance indicators (i.e. pressure drop). Therefore, an automatic water flow control system is essential for systematic biofouling studies in MFSs.



Fig. 4. Feedwater flow rate (a) and pressure drop in time (b) over the MFS systems with automatic flow control fed with water without (AS-) and with dosed (AS) biodegradable substrate.



Fig. 5. Pressure drop in time (a) and pressure drop increase (b), and accumulated biomass in ATP and TOC (c,d) for manually and automatic controlled setups. Manual flow control without substrate (MS-) and with dosed substrate (MS). Automatic flow control without substrate (AS-) and with dosed substrate (AS). In all cases, the initial pressure drop was lower for the automatic setups.

4.1. Importance of accurate and stable feed flow

A suite of monitors has been developed to study the biofilm formation and related performance decline (biofouling) in membrane systems for water treatment [31]. For representative biofouling studies, the MFS units are designed to have similar hydrodynamics as in spiral wound membrane modules. Although the developed MFSs are suitable for biofouling studies, the way in which the monitors are operated may impact the results. Hydrodynamic conditions affect biofilm formation and morphology in spiral wound membrane systems [11,32]; at lower flow rates: (i) the same amount of accumulated biomass has a lower impact on membrane performance and (ii) biofilm accumulation is slower, while at high flow rates biofilm accumulation is more rapid and a different biofilm morphology is observed. The water flow rate and thus, water flow rate variations have an impact on the biofilm accumulation rate and morphology and changes in flow rate may lead to biomass detachment. The impact of accumulated biomass on the feed channel pressure drop depends on the linear flow velocity [11,33]. Clearly, constant and accurate water flow is very important.

The developed automatic system enabled a constant flow rate with variations smaller than 1% throughout the experimental studies, while for the manual system variations were found up to 3% without biofilm formation and up to 9% with biofilm formation. The manual system biofilm formation strongly influenced the flow rate.

Membrane performance is evaluated by feed channel pressure drop increase and/or permeate flux decline. Both membrane performance parameters are affected by the water flow rate. Our study illustrates that a sensitive, accurate water flow monitoring is required for an accurate determination of the impact of fouling accumulation on membrane performance parameters. In summary, stable and accurate feedwater flow rates are needed for controlled membrane biofouling studies using monitors. The developed automatic setup for monitor operation is suitable to maintain an accurate constant water flow rate enabling reliable pressure drop measurements.

4.2. Evaluation of earlier research done with manual flow control

Earlier membrane biofouling studies with monitors used a manual flow control system involving a combined flow control with back pressure valve which was located after the monitor (Fig. 1(a)). The flow control system had a spring and membrane regulating the water flow to a certain extent.

The manual water flow system showed the same accumulated amount of biofilm as the automatic flow system after the 9 d study, in which the feed channel pressure drop over the monitors showed a clear difference in pressure drop increase (Fig. 5). The same biofilm accumulation illustrates that the results of the earlier membrane biofouling studies using MFSs with manual flow control are still valid. However, systematic well-controlled studies on biofouling require a stable water flow.

5. Conclusions

An automatic system for constant and automatic flow control of feedwater to MFSs was developed and

evaluated. Results were compared with a manual flow control system, previously used for membrane biofouling studies. Based on the results, the following conclusions can be made:

- (1) Stable and accurate feedwater flow rates are needed for controlled membrane biofouling studies: this was achieved by the automatic setup.
- (2) Correct pressure drop measurements requires an accurate constant water flow rate.
- (3) The developed automatic system for monitor operation is suitable to maintain an accurate constant water flow rate, enabling accurate pressure drop measurements and more stable biofilm growth.

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