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# Biofouling removal in spiral-wound nanofiltration elements using two-phase flow cleaning

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## ABSTRACT

Biofouling has detrimental effects on the feed channel pressure drop and the permeate flux in high-pressure membrane processes such as NF and RO. Two-phase flow cleaning is a chemical-free technique that is able to remove such biofilms. This paper presents a study into the effects of the gas/liquid ratio, feed spacer geometry, applied pressure and liquid velocity on the efficiency of two-phase flow cleaning in spiral-wound nanofiltration elements. A high-speed camera, optical coherence tomography and scanning electron microscopy were used to study biofouling and its removal. Our results show that two conditions must be met to ensure that a sufficiently high shear force is applied to biofilms on membrane and spacer surfaces. A good bubble distribution in the channel is the first requirement. While it is mainly the structure of the feed spacer that controls bubble flow and bubble size, a minimum gas/liquid ratio of 0.5 is necessary to achieve a good bubble distribution. The second condition is the use of a sufficiently high liquid velocity during cleaning. The bubble velocity was found to be 3.5–5.5 times as high as the used liquid velocity, and responsible for a marked improvement in the flux recovery.

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## 1. Introduction

Biofouling is considered the biggest vulnerability of high-pressure membrane processes used for water purification, such as nanofiltration (NF) and reverse osmosis (RO). Biofouling often flourishes in filtration media and membrane systems, but inside water distribution pipes, it also poses a problem [1]. Biofouling results in operational problems such as a rapid increase of the feed channel pressure drop leading to a flux decline, and quickly reoccurs after chemical cleaning [2–4].

Biofouling in membrane processes is dominated by bacteria living in surface-associated multicellular communities known as a biofilm [5,6]. The bacteria in these biofilms are surrounded by extracellular polymeric substances (mainly polysaccharides, proteins, nucleic acids and lipids) [7]. Microorganisms are not the only cause of biofouling in membrane processes, however. Water also contains organic compounds that are for instance released into the water during an algal bloom (extracellular organic matter or EOM) or are derived from dead biomass, for example resulting from the use of biocides (autochthonous organic matter or AOM). These organic compounds

constitute a nonliving form of biofouling and aggravate microbiological fouling [8,9].

Traditionally, two different strategies are used to mitigate biofouling [10]. The first approach is to remove microorganisms before they enter an RO or NF system. Such a pretreatment stage (e.g. sand filtration, ultrafiltration) can be crucial to improve the quality of feed intake as it lowers the silt density index, removes algae, molds and bacteria, and decreases the concentration of total and dissolved organic carbon [11–13]. According to a market analysis, ultrafiltration pretreatment is gaining popularity in favor of conventional pretreatment, the installed ultrafiltration pretreatment capacity having exceeded 1,000,000 m<sup>3</sup>/d in 2008 [14]. However, ultrafiltration does not provide total protection against many dissolved organic compounds (notably polysaccharides and transparent exopolymer particles), and some biofouling in NF and RO system is therefore unavoidable.

The second approach is to deactivate (or kill) microorganisms chemically during a so-called “cleaning in place” or CIP treatment. Different types of chemical cleaning agents can be applied (e.g. alkaline, acids, biocides, detergents, enzymes *etc.*), usually suggested by the membrane manufacturer. Chemical cleaning, however, does not guarantee a flux recovery of 100%, since it does not remove any biomass, but only deactivates it [15]. The remaining biomass will still cause operational problems, and acts as a substrate for newly

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attached bacteria [16]. The use of chemical cleaning agents also causes a waste problem, and frequent cleaning with aggressive agents contributes to loss of membrane integrity resulting in shorter lifetimes and an increase of operational costs [17].

A detailed study of the effects of conventional chemical treatment on the initiation and spatiotemporal development of biofouling (during short-term and long-term operation in an RO water purification plant) revealed that state-of-the-art cleaning-in-place by chemicals failed to control biofouling [18]. A combination of molecular (FISH, DGGE, clone libraries and sequencing) and microscopic (FESEM, CLSM) analyses showed that bacterial recolonization of the biofilm layers disrupted by chemical treatment starts directly after chemical cleaning by attachment and growth of primary colonizers from the intakes, and by proliferation of microorganisms that survived the chemical cleaning within the collapsed biofilm layer [18]. Microbiological studies by Costerton et al. [19] elucidated that bacteria living in such nutrient-sufficient environments are hundreds of times more resistant to antibacterial agents. Removal of all (mostly dead) biomass after a chemical cleaning is expected to prevent this rapid re-growth of biofilms [18].

Controlling the hydrodynamics around a biofilm is crucial for its disruption and detachment from surfaces [20,21]. A novel low-cost chemical-free method for biofouling removal from membrane systems involves the use of a two-phase flow cleaning technique [22]. Two-phase flow cleaning is able to remove biofouling in spiral-wound modules, as demonstrated by a decrease in the feed channel pressure drop [23,24]. Nevertheless, a better understanding of the underlying mechanisms of two-phase flow cleaning in various operational settings can help improve its cleaning efficiency. In this paper, findings obtained during a study into the development of biofouling in spacer-filled rectangular flow cells simulating spiral-wound nanofiltration elements and its removal by two-phase flow cleaning are reported.

In order to understand the development and removal of biofilms, it is important to study their development and removal in time as well as in space. The initial attachment of a biofilm occurs within minutes to hours [25], but the subsequent growth, detachment, re-growth and maturation times can be in the order of days, weeks, months and years. This depends on the diversity of the microbial consortia in the biofilm, nutrient inhibitors, hydrodynamics, and the geometrical characteristics of the surface to which the microorganisms are adhered [26]. These time scales are also related to a three-dimensional structure. First of all, the biofilm is heterogeneously distributed within spiral-wound membrane elements. Our scope of interest concerns three spatial scales: biofilm development and distribution in the elements including distribution of two-phase flow during biofilm removal (macroscale), the heterogeneity of a biofilm on the membrane and feed spacer surfaces (mesoscale), and observation of constituents of microbial colonies (microscale). The microscale usually is characterized by the distance between two microbial cells (1–10  $\mu\text{m}$ ), while the mesoscale is defined by the average biofilm thickness (10–1000  $\mu\text{m}$ ) [27,28]. Both *in-situ* and *ex-situ* inspections of fouled membrane and spacers through the use of a high-speed camera, optical coherence tomography (OCT) and scanning electron microscopy (SEM) therefore were conducted. This kind of knowledge is essential to be able to draw up guidelines for optimal biofouling removal using a two-phase flow cleaning technique and provide more insight regarding its practical applicability in large-scale installations.

## 2. Theory

### 2.1. Increase of the feed channel pressure drop (FCP)

Two types of pressure differences can be distinguished in membrane systems: the feed channel pressure drop (FCP) and

the trans-membrane pressure drop or difference, also called trans-membrane pressure (TMP). The TMP results from the feed pressure and is the pressure difference between the feed side and the permeate side, applied to overcome the total resistance across the membrane. The FCP is the pressure difference between channel inlet and channel outlet due to the hydraulic resistance of the channel. The FCP is commonly described in terms of the dimensionless friction factor ( $\lambda$ ) and the Reynolds number (Re). The empirical constants relating these two parameters depend on the characteristics of the feed channel and on the flow type (laminar or turbulent) of the fluid in the channel. In spiral-wound elements, the latter is determined by the type of feed spacer used (commonly diamond-mesh extruded netting).

The FCP in spacer-filled channels can be expressed by the following equations [29]:

$$\text{FCP} = \Delta P = \lambda \cdot \frac{\rho u^2}{2} \cdot \frac{L}{d_h} \quad (1)$$

$$\lambda = 6.23 \text{Re}^{-0.3} \quad (2)$$

$$\text{Re} = \frac{\rho \cdot d_h \cdot u}{\mu} \quad (3)$$

where  $\Delta P$  is the pressure drop (Pa),  $\rho$  is the liquid density ( $\text{kg/m}^3$ ),  $d_h$  is the hydraulic diameter of the channel (m),  $u$  is the specific liquid velocity (m/s),  $\mu$  is the liquid dynamic viscosity (Pa s) and  $L$  is the length of the spacer-filled channel (m). Eq. (2) is valid for  $100 < \text{Re} < 1000$  and all types of feed spacer.

The presence of feed spacers reduces the porosity ( $\varepsilon$ ) or void volume in the channel, resulting in a higher specific velocity when compared with an empty channel, as follows:

$$u = \frac{\phi}{\varepsilon \cdot W \cdot H} \quad (4)$$

where  $\phi$  is the volumetric flow rate ( $\text{m}^3/\text{s}$ ), and  $W$  and  $H$  are the feed channel width and height, respectively (m). The porosity of the feed spacer and hydraulic diameter are estimated by using the following equations [29]:

$$\varepsilon = 1 - \frac{V_{sp}}{V_{tot}} \quad (5)$$

$$d_h = \frac{4 \cdot \varepsilon}{(2/H) + (1 - \varepsilon) \cdot (4/d_f)} \quad (6)$$

where  $V_{sp}$  is the spacer volume ( $\text{m}^3$ ) and  $V_{tot}$  the total volume of the feed channel ( $\text{m}^3$ ).

In the case of a diamond-shaped spacer with a  $90^\circ$  angle, such as used in spiral-wound elements, the following two equations from Schock and Miquel [29] can be used:

$$V_{sp} = \frac{\pi}{2} d_f^2 L_f \quad (7)$$

$$V_{tot} = H L_f^2 \quad (8)$$

in which  $d_f$  is the diameter of the feed spacer filament (m), and  $L_f$  is the length of the feed spacer filament (m).

As the biofilm attaches and grows in the spacer-filled channel, the channel's porosity and hydraulic diameter decrease which results in an increase in the specific velocity and the FCP. This strongly affects the flow distribution. The increased pressure drop in the feed channel also leads to a reduction of the trans-membrane pressure, hence reducing permeability. A typical threshold above which operational problems occur in industrial systems is an increase of 15% of the FCP over the entire installation [16].

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