



# The composition and compression of biofilms developed on ultrafiltration membranes determine hydraulic biofilm resistance



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## ABSTRACT

This study aimed at identifying how to improve the level of permeate flux stabilisation during gravity-driven membrane filtration without control of biofilm formation. The focus was therefore on understanding (i) how the different fractions of the biofilms (inorganics particles, bacterial cells, EPS matrix) influence its hydraulic resistance and (ii) how the compression of biofilms impacts its hydraulic resistance, i.e., can water head be increased to increase the level of permeate flux stabilisation. Biofilms were developed on ultrafiltration membranes at 88 and 284 cm water heads with dead-end filtration for around 50 days. A larger water head resulted in a smaller biofilm permeability (150 and 50 L m<sup>-2</sup> h<sup>-1</sup> bar<sup>-1</sup> for biofilms grown at 88 cm and 284 cm water head, respectively). Biofilms were mainly composed of EPS (>90% in volume). The comparison of the hydraulic resistances of biofilms to model fouling layers indicated that most of the hydraulic resistance is due to the EPS matrix. The compressibility of the biofilm was also evaluated by subjecting the biofilms to short-term (few minutes) and long-term variations of transmembrane pressures (TMP). A sudden change of TMP resulted in an instantaneous and reversible change of biofilm hydraulic resistance. A long-term change of TMP induced a slow change in the biofilm hydraulic resistance. Our results demonstrate that the response of biofilms to a TMP change has two components: an immediate variation of resistance (due to compression/relaxation) and a long-term response (linked to biofilm adaptation/growth). Our results provide relevant information about the relationship between the operating conditions in terms of TMP, the biofilm structure and composition and the resulting biofilm hydraulic resistance. These findings have practical implications for a broad range of membrane systems.

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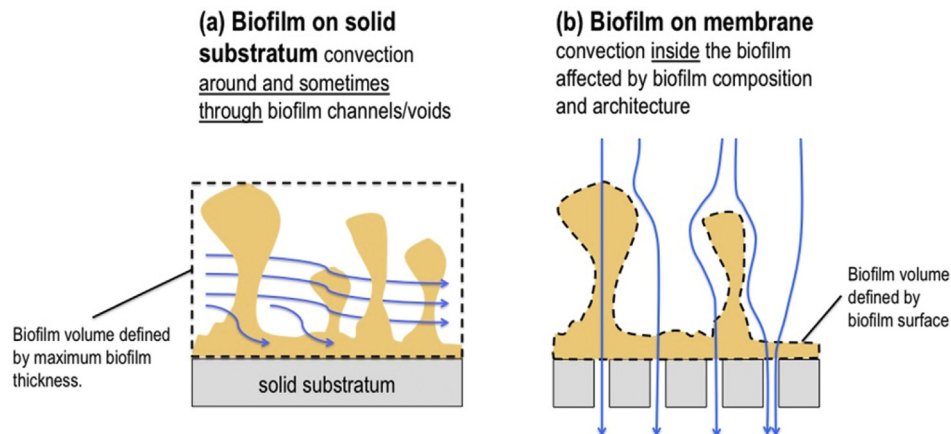
## 1. Introduction

Biofilms inevitably grow on membrane surfaces and reduce permeate flux. So far the operation of membrane systems mainly relied on avoiding biofilm formation. Different strategies were developed to control the biofilm growth. But recent studies suggested that it might be possible or desirable to live with biofilm (Derlon et al., 2014; Dreszer et al., 2013). Biofilm-membrane composite system indeed have multiple advantages compared to membrane system only – flux stabilisation, improved permeate

quality. Whatever the selected approach (living with or fighting the biofilms), it is key to understand what factors determine the hydraulic resistance of biofilms. This is especially relevant for membrane systems where biofilm formation is fully tolerated (Derlon et al., submitted) or controlled to a low extent (Smith et al., 2015).

Biofilms are dynamic and complex structures made of different organic (e.g., cells, EPS) and inorganic fractions. Mass transport of soluble substrates and fluid dynamic outside of the biofilm is rather well understood. But very little information about the water flow through the biofilm itself is available. Convection through biofilms has been studied for biofilms developed on solid substrata under cross-flow conditions (de Beer et al., 1996; Lewandowski et al., 1995; Stoodley et al., 1994). Convection around and sometimes through cell clusters was observed using fluorescein or fluorescent particles (de Beer et al., 1994; Stoodley et al., 1994) (Fig. 1a). A

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**Fig. 1.** Conceptual representations of convection through (a) Conceptual representation of convection through biofilms grown on solid substrata. The flow occurs here mainly between the biofilm heterogeneities and sometimes beads penetrate the biofilm matrix. (b) Conceptual representation of convection through biofilms grown on permeable substrata such as membranes, i.e., the main research question addressed in this study.

similar observation was reported for heterogeneous biofilms developed in porous media, for which advection is often observed around heterogeneities (Flemming et al., 2000). These studies were then extensively cited and it became well accepted that convection occurs inside the biofilms. However, channels in the studies of Stoodley et al. (1994) and de Beer et al. (1994) refer to external voids, i.e., to the valleys or conduits separating cells clusters, streamers or other structural heterogeneities. Thus, it still remains unclear whether convection occurs inside the volume of the biofilms that is defined by the biofilm-bulk interface vs. convection inside the volume defined by the maximum biofilm thickness as observed in the studies of Stoodley et al. (1994).

Understanding what are the factors that influence the hydraulic resistance of biofilms is particularly important for membrane biofilms (Fig. 1b). It is especially important to identify how the composition and internal architecture of the biofilms may influence permeation (e.g., presence of internal voids/channels, cells, EPS matrix) (Fig. 1b). The comparison of the hydraulic resistances of biofilms with the one of model fouling layers gave initial insights about the relationship between composition and hydraulic resistance. Different studies suggested that EPS might be the main contributor to biofilm hydraulic resistance (Dreszer et al., 2013; McDonogh et al., 1994; Stewart, 2012). Stewart (2012) compared the permeability of model fouling layers made of spheres or hydrated gels. The layers made of spheres were more permeable than the layers of hydrated gels. Stewart (2012) thus concluded that the EPS content of biofilms governs the biofilm permeability. Dreszer et al. (2013) compared the overall resistances of bacterial cell layer and biofilms (containing the same amount of cells). The overall resistance of the cell layer was significantly smaller than the overall resistance of a biofilm that contained the same volume of cells (6-fold difference). Dreszer et al. (2013) thus attributed the difference in the hydraulic resistances to the EPS. However the mass/thickness of the cell layer was much lower than those of the biofilm. The difference between the specific resistances (resistance relative to mass or thickness) is less pronounced than the one of the absolute resistances (3-fold vs. 6-fold, respectively). The work of Stewart (2012) and Dreszer et al. (2014) thus provide plausible insights about the influence of the EPS on the biofilm permeability. But other studies reported contradictory findings, i.e., that bacterial cells are more resistant to permeation than EPS (McDonogh et al., 1994). Thus, it is still required to evaluate how the different biofilm fractions (inorganic particles, cells and EPS) impact its hydraulic resistance.

Better understanding how biofilm mechanics (e.g., compression) influence the biofilm permeability is also an important aspect of membrane biofilms. It is intuitive that the biofilm composition likely determines the mechanical properties of the biofilms, i.e., how biofilms respond to stresses and ultimately change their internal structure and permeability. Biofilms grown on solid substrata under cross-flow conditions behave as viscoelastic material (Stoodley et al., 1999a, 1999b). The strain increases linearly at low load (elastic response) and then a creep is observed over time (viscoelastic response). Studies that applied a normal force also showed that biofilms are compressible. Pure culture biofilms from the dental pathogen *Streptococcus mutans* were for example highly compressible when applying a normal force of 0.1 N over a 25 mm diameter disk (Vinogradov et al., 2006). However, very little is known about the compressibility of biofilms growing on permeable substratum such as membranes. Young biofilms grown on membrane surfaces with acetate-based feed solutions were shown to be compressible when increasing the permeate flux from 20 to 60 L m<sup>-2</sup> h<sup>-1</sup> (Dreszer et al., 2014). The study of Dreszer et al. (2014) delivered very relevant insights about the effect of an increased TMP on biofilm compressibility and resistance. But the compressibility of biofilms must also be evaluated for older biofilms characterised by a more complex composition.

This study aims at better understanding (i) how the biofilm composition (inorganic fraction, cells, EPS) influences its hydraulic resistance and (ii) how the mechanical properties of the biofilms (compressibility) ultimately influences its hydraulic resistance. Biofilms were developed during gravity-driven membrane ultra-filtration at two different water heads: 88 and 284 cm. Permeate flux and permeability were analysed with regard of the biofilm composition. Biofilm composition was characterised in terms of inorganic and organic carbon (bacterial cells, EPS) concentrations and volumes. Biofilms of different ages were then submitted to short- and long-term step-wise increase of TMP and to evaluate their compressibility.

## 2. Materials and methods

### 2.1. Operating conditions

Two types of experiments were performed in this study: (1) long-term filtration experiments with biofilm formation on membrane surfaces at a constant pressure for several weeks (Exp. 1.1, Table 1 first row) or at constant pressure during an initial growth

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