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Fouling mechanism elucidation in membrane bioreactors by bespoke physical cleaning



G. Di Bella^{a,*}, D. Di Trapani^b, S. Judd^c

- ^a Facoltà di Ingegneria e Architettura Università degli Studi di Enna "Kore", Cittadella Universitaria, 94100 Enna, Italy
- b Dipartimento di Ingegneria Civile, Ambientale, Aerospaziale, dei Materiali, Università degli Studi di Palermo, Viale delle Scienze, 90128 Palermo, Italy
- ^c Cranfield Water Science Institute, Cranfield University, Bedfordshire MK43 0AL, United Kingdom

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ABSTRACT

Cake layer deposition on a membrane surface can determine both external and internal membrane fouling through negatively affecting the total filtration resistance while exerting a positive effect as a pre-filter. Membranes are usually subjected to a periodic cake layer removal through routine physical cleaning, specifically permeate backwashing of hollow fiber membranes, or enhanced cleaning through, for example, chemically-enhanced backwashing. Physical cake layer removal is crucial for sustaining permeability, yet the effect of different physical cleaning modes remains poorly evaluated. The present work attempts to analyze physical cake layer removal through the application of specific cleaning methods and the impact of these on the subsequent resistance. The constituent contributions to the overall resistance are appraised by means of the Resistances In-Series model, with the aim of producing a robust protocol for quantifying these discrete contributors. The results, based in part on published data, show the proposed approach to reliably determine the relative contribution of the different resistance components to within $0.1 \cdot 10^{12} \, \mathrm{m}^{-1}$ across a range of different bench and pilot-scale plants, confirming the resilience of the method.

1. Introduction

Physical and chemical cleaning of membrane bioreactors (MBRs) to sustain permeability represents a crucial component of MBR operation [1]. Physical cleaning notionally removes the loosely attached material on membrane surfaces, usually referred to as "reversible fouling", whereas the more aggressive chemical cleaning removes more tenacious materials, or "irreversible fouling" [2]. Reported improvements in sustaining permeability through physical means have included water washing [3,4], ultrasonic cleaning [5], high-frequency vibration [6], and the use of ancillary particles for in-situ mechanical cleaning [7–9].

Notwithstanding these developments, the factors determining the efficacy of membrane physical cleaning have not yet been fully discerned, primarily because of the widely-acknowledged complexity of fouling itself [1,10–12]. In particular, the role of the cake layer, and specifically how it pertains to reversible and irreversible fouling, remains contentious. According to a number of authors [13–15], a proportion of the cake layer deposited on the membrane surface can be considered "irreversible" if only removable by *enhanced* physical methods, such as ex-situ water flushing, mechanical cleaning, or cyclic cleaning – e.g. combined backwashing and crossflushing [13]. There

should therefore be a distinction between reversible fouling removable by conventional backwashing and relaxation and that removed by the enhanced physical methods. This distinction is of practical significance, since enhanced physical methods may be preferred to the application of chemicals for recovering permeability.

Physical washing of MBR membranes in experimental studies is usually employed for cleaning purposes, rather than for elucidation of fouling mechanisms. Few authors have used data derived from periodic physical cleaning for the analysis of fouling development and defining deposition mechanisms. In the latter context, the Resistance-In-Series (RIS) model represents one of the most extensively used approaches [16,17] since it is intuitive and allows quantification of the discrete fouling components, albeit with some limitations [18,19]. The use of physical cleaning to define fouling components as defined by the RIS model has nonetheless been limited.

The aim of the present study is to gain insight into the usefulness of membrane physical cleaning for detailed analysis of fouling mechanisms by delineating the different components using the RIS model. The RIS model has been applied to the outputs from a protocol encompassing manual physical washing, and their reproducibility subsequently determined. The model was applied to outputs from previous

E-mail addresses: gaetano.dibella@unikore.it (G. Di Bella), daniele.ditrapani@unipa.it (D. Di Trapani), s.j.judd@cranfield.ac.uk (S. Judd).

^{*} Corresponding author.

studies [20–24] pertaining to a range of pilot and bench scale plants of different configurations, operational conditions and feed wastewater characteristics. The system behavior in terms of removal efficiency, biomass activity and fouling propensity has been analyzed. The fouling mechanisms and development were re-evaluated using a standard computational approach to define new specific resistance values, as well validating simulated data under the range of operating conditions employed in the pilot-scale installations. Supplementary specific batch tests were performed under comparable conditions using similar membrane modules to assess the robustness of the adopted protocol with reference to the precision of the outputs.

2. Materials and methods

2.1. Protocol and RIS model

A bespoke physical cleaning protocol was employed to define the characteristic resistances associated with the main fouling mechanisms. The protocol was reproduced by the operator for all the plants subjected to physical cleaning at least once during the plant operation. The permeate flux and transmembrane pressures (TMP) were measured during normal plant operations, prior to cleaning, the total resistance to filtration ($R_{tot.1}$) being defined as:

$$R_{tot,1} = \frac{TMP_1}{J_1 \cdot \mu} \tag{1}$$

where J_1 is the permeate flux of the fouled membrane [m³ m⁻² s⁻¹], TMP_1 the transmembrane pressure [Pa], μ the permeate viscosity [Pa s] at the operating temperature.

The membrane was then removed from the bioreactor and physically cleaned by (a) rinsing with tap water at 0.4–0.5 bar for 15 min with mild mechanical cleaning, (b) mechanical agitation in water for 5 min, and (c) rinsing with ultrapure water at < 0.2 bar for a further 5 min. The cleaned membrane was then immersed in clean water and subjected to a normal filtration cycle (with the same operational flux, and ultimately with conventional backwashing if appropriate) to allow measurement of the resistance to filtration in clean water ($R_{tot.cw}$):

$$R_{tot,cw} = \frac{TMP_{cw}}{J_{cw} \cdot \mu} \tag{2}$$

where J_{cw} and TMP_{cw} are the permeate flux and the TMP in clean water after physical cleaning, respectively.

The membrane was then placed back in the bioreactor and subjected to the normal filtration cycle (with the same flux values and classical backwashing), using the same mixed liquor as in Step 1, to evaluate the final total resistance to filtration ($R_{tot,2}$):

$$R_{tot,2} = \frac{TMP_2}{J_2 \cdot \mu} \tag{3}$$

where J_2 and TMP_2 refer to physically-cleaned membrane conditions. Thus, the total resistance $R_{tot,2}$ can be expressed as the sum of the intrinsic membrane resistance (R_m) , the resistance due to pore blocking (R_{PB}) and the resistance due to reversible cake deposition $(R_{C,rev})$, since the irreversible cake is not formed at the first cycle after physical cleaning. The total resistance $R_{tot,1}$ during normal operation can be expressed as:

$$R_{tot,1} = R_m + R_{PB} + R_{C,rev} + R_{C,irr}$$
(4)

where R_m and R_{PB} represent the membrane and the pore blocking resistance contributions. In contrast, the resistances $R_{tot,cw}$ and $R_{tot,2}$ are given by:

$$R_{tot,cw} = R_m + R_{PB} \tag{5}$$

$$R_{tot,2} = R_m + R_{PB} + R_{C,rev} \tag{6}$$

where

$$R_{PB} = R_{tot,cw} - R_m \tag{7}$$

$$R_{C,irr} = R_{tot,1} - R_{tot,2} \tag{8}$$

$$R_{C,rev} = R_{tot,2} - R_{tot,cw} \tag{9}$$

The cake resistance, either reversible $(R_{c,rev})$ or irreversible $(R_{c,irr})$, is considered to be completely removable by physical cleaning. The superficial fraction removable by backwashing and air scouring relates to the less tenacious cake layer $(R_{c,rev})$, while the fraction removable by manual washing is the more tenacious superficial cake layer $(R_{c,irr})$. The residual fouling layer, which is not removed by either backwashing or physical washing, pertains to intermediate pore blocking. Indeed, it was supposed that the removal of pore blocking fraction by physical operations (both backwashing and physical cleaning) is negligible if not supported by the addition of chemicals.

2.2. Investigated plants

Resistance data derived from previous studies, the main findings of which have been published elsewhere, were reprocessed to combine the outputs from the physical cleaning with the RIS model application for both hollow fibers (HF) and flat sheet (FS) membrane modules. Most of the results refer to $\sim 0.1~\text{m}^2$ membrane area bench scale HF modules scoured at aeration rate of $0.6~\text{Nm}^3~\text{m}^{-2}~\text{h}^{-1}$. Under these conditions, the system hydrodynamics are widely known not to reflect those of full-scale modules due to the disproportionate impact of the headers [25] and the conflict between the specific aeration demand and the average air upflow velocity [26]. In the present study, new filtration batch tests were conducted on mixed liquor of known characteristics to obtain reliable and reproducible results, along with tests on larger, pilot-scale $0.93~\text{m}^2$ HF modules air-scoured at rates of $0.5~\text{Nm}^3~\text{m}^{-2}~\text{h}^{-1}$ and $0.8~\text{m}^2$ FS modules scoured at $0.75~\text{Nm}^3~\text{m}^{-2}~\text{h}^{-1}$ (Table 1).

The bench or pilot plants listed in Table 1 were operated in continuous mode and the membrane modules subjected to ex-situ manual washing only when either the TMP reached a given threshold value suggested by the manufacturer or the operational conditions of the plant were expressly changed. These data refer to single washing operations and to the conditions prior to and immediately after physical cleaning (Eqs. (1)(9)). The proposed approach then enabled a comparison of the fouling tendency as well elucidation of the main fouling mechanisms of the different MBR systems and quantifying the impact of the enhanced physical cleaning. Recorded data include mixed liquor composition - the suspended solids (SS), extracellular polymeric substances (EPSs) and salinity - along with the values for the total resistance (Rtot), and the discrete resistance values associated with pore blocking (R_{PB}) and the reversible and irreversible cake layer $(R_{C,rev})$ and $R_{C.irr}$). The RIS model was applied during the physical washing employed during normal pilot plant operation.

Physical cleaning of either bench or pilot scale plants was carried out following filtration periods characterized by the same operating conditions at pseudo-steady state. Key recorded parameters comprised:

- average mixed liquor suspended solid (MLSS) concentration, relating to reversible superficial deposition (i.e. removable by backwashing);
- <u>average specific EPS concentration</u> in the mixed liquor (per gram of SS), relating to irreversible (internal and superficial) fouling;
- mean filtration period, the elapsed time t₀ between previous physical cleaning, prolonged filtration directly influencing superficial and/or internal irreversible fouling;
- instantaneous permeate flux.

The investigated bench or pilot plants were fed with real municipal wastewater [20,21,27,28], high strength synthetic wastewater characterized by a sharp salinity increase [22] and synthetic wastewater characterized by a gradual salinity increase [23].

Concerning the plant MBR_1 , the protocol was applied twice and two

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