Chapter

Fouling Monitoring in Membrane Filtration Systems

Luca Fortunato

Abstract

Membrane filtration systems are employed in the water industry to produce drinking water and for advanced wastewater treatment. Fouling is considered the main problem in membrane filtration systems. Fouling occurs when the biomass deposited on the membrane surface leads to a membrane performance decline. Most of the available techniques for characterization of fouling involve the analysis of membrane samples after membrane autopsies. This approach provides information ex-situ destructively at the end of the filtration process. Optical coherence tomography (OCT) gained attention in the last years as noninvasive imaging technique, capable of acquiring scans in-situ and nondestructively. The online OCT monitoring enables visualizing and studying the biomass deposition over time under continuous operation. This approach allows to relate the impact of the fouling on the process. In the last years, the suitability of OCT as in-situ and nondestructive tool for the study of fouling in membrane filtration systems has been evaluated. The OCT has been employed to study the fouling in different membrane geometry and configuration for the treatment of seawater and wastewater. Nowadays, the OCT is employed to better understand the role of biomass structure on the filtration mechanisms.

Keywords: OCT, water treatment, desalination, biofouling, fouling, membrane filtration

1. Water scarcity

Nowadays, the insufficiency of access to clean and secure water represents one of the main problems for sustainable development affecting both industrialized and developing countries. It is estimated that by 2030, almost 50% of the global population will face water stress conditions. Although 71% of the planet Earth's surface is covered by water, only less than 3% is constituted by fresh water, where the remaining 97% of the water is seawater and characterized by the high content of mineral salt, which makes it inadequate for direct consumption.

The continuous increase in the human population and industrialization lead to a constant increase of the water demand. Freshwater is not used only for direct consumption, but it is the central pillar of food and energy production. The use of water in agriculture contributes to 70% of the total water withdrawn [1]; the other major contribution to the global consumption is related to the energy production, whereas water is required in the whole cycle of the energy production and distribution. Besides the scarcity, another major threat is represented by the quality of the water available. According to the UNESCO, almost 3.6 billion people, lack for access to clean water and proper sanitation [2]. To meet the continuous demand, there is a need to produce freshwater starting by nonconventional sources, as saline water (seawater and brackish) and other contaminated fresh water sources (wastewater and industrial water). Among the different technologies tested over the years, membrane filtration is recognized as the most common and convenient method to purify water.

1.1 Membrane technology

The membrane is a semi-permeable barrier used to separate specific substances considered as pollutants from the water. The purification occurs by applying pressure in the systems, whereas the membrane allows only certain ions and molecules to be transported through the membrane with a specific size to pass, retaining all the rest. During the filtration, the water passes through the membrane and the rejection of other compounds depends on the pressure applied and the membrane pore size. The main membrane filtration processes and their respective removal capabilities are shown in **Figure 1**. By using different membranes, it is possible to remove specific unwanted compounds. Hence, safe drinking water can be produced starting from different sources, including seawater and wastewater.

Membranes can be classified according to different criteria. The most diffused classification is based on the membrane pore size, whereas the removal of unwanted contaminants is related to the membrane pore size. The highest removal is realized by reverse osmosis membranes (RO) followed by nanofiltration membrane (NF), are generally used to remove salt from water (desalination from seawater). Ultrafiltration (UF) and microfiltration (MF) membranes, with pore size from 0.001 to 0.1 μm, are employed to remove pathogens and suspended solids. RO membrane requires high pressure up to 70 bar to remove the salt during desalination processes, while MF and UF usually require lower pressure less than 5 bar [3].

Another classification is based on the geometry of the membrane employed in the process (i.e., flat sheet, spiral wound, tubular, and hollow fiber). The use and choice of a specific geometry are often linked to a specific application and system design,

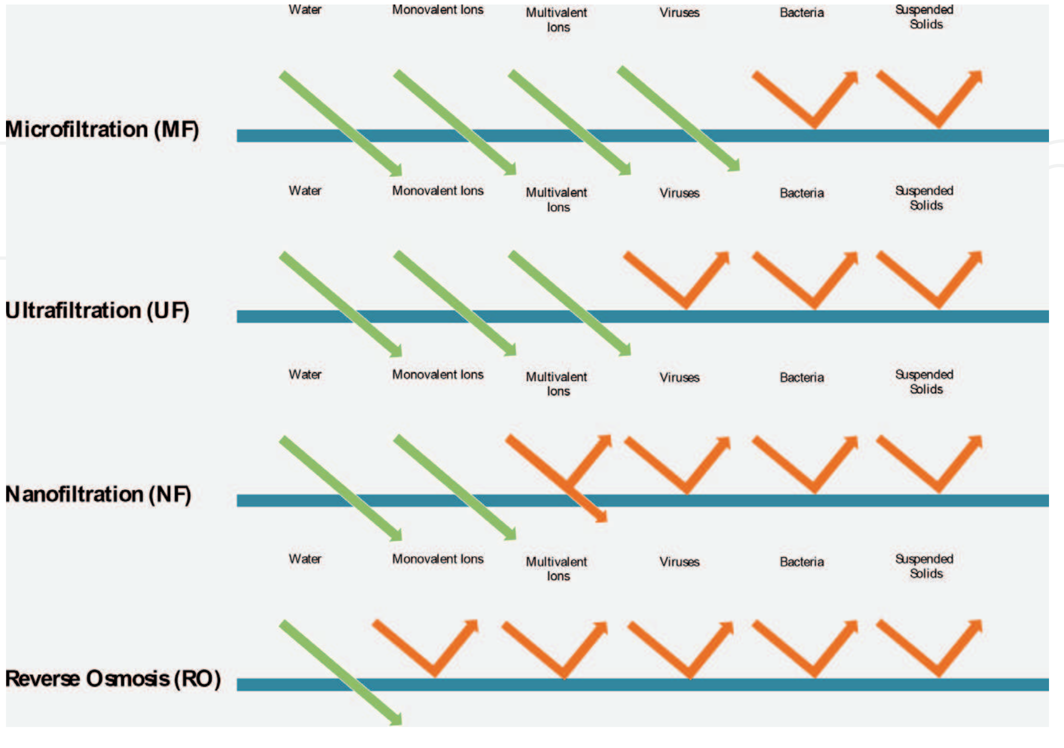

Figure 1.
Membrane size and compounds separation.

where the membrane module can be submerged or external. The most employed submerged membrane configuration is the membrane bioreactor (MBR), whereas the membrane unit can be directly submerged in the aeration tank or in a separate tank. On the contrary, commercial seawater membrane desalination plants employ RO membrane as external module inserted in vessels that allows the use of high pressure (60–70 bar) necessary to remove salt from water. These membranes are called spiral wound membrane modules, and consist of membrane sheets enfolded along a central tube, comprising a perforated central tube for permeate collection surrounded by layers of membrane, permeate spacers and feed spacers. The produced water, called permeate, is collected in the central tube by the product spacer.

1.2 Membrane fouling

The continuous filtration of water over time leads to the accumulation of rejected material on the membrane surface and or in membrane pores. This phenomenon is called fouling. The development of membrane fouling on the membrane surface is considered the bottleneck of membrane filtration processes. Over time, fouling is inevitable and leads to the decrease of the membrane flux that is considered as the main process performance indicator. The decrease in flux is followed by the energy increase as the pressure applied to overcome the reduction in water production. Moreover, to recover the membrane permeability, the operators increase the use of chemicals for the cleaning, therefore reducing the membrane lifetime. Therefore, the fouling understanding and control are considered the major challenges encountered in membrane filtration processes as highlighted by the crescent number of scientific publications and the increase of commercial products for the reduction and control of fouling.

The fouling deposited in membrane filtration systems varies depending on each specific process, and it is due to the complex interaction between the constituents present in the feed water and the membrane [4, 5]. The fouling mechanism generally involves (i) initial pore blocking followed by (ii) cake layer formation. Depending on the characteristics of the water treated, the fouling in membrane filtration systems can be divided into four categories (**Figure 2**):

- Particulate fouling
- Organic fouling
- Inorganic fouling (or scaling)
- Biofouling

Particulate fouling is due to the accumulation of suspended particles of different nature on the membrane surface. Organic fouling consists of the formation of an organic layer on the surface and is linked to the concentration of dissolved organic

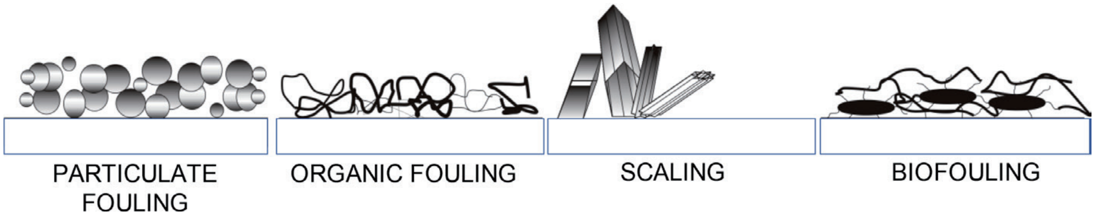

Figure 2.
Different types of fouling deposited on the membrane surface.

compounds in the feed water. The scaling is the most common type of inorganic fouling in desalination processes; it is due by the direct formation of crystals on the membrane surface through precipitation. Depending on the process, different types of fouling can occur concurrently; in that case, often the fouling deposited on the membrane is referred as biomass.

Among the different types of fouling membrane, biofouling is considered the most problematic fouling faced in membrane filtration system, which negatively affects the process in terms of technology and economics [6]. Biofouling refers to the development of biofilms in the membrane systems and it is caused by the accumulation of microorganisms, including extracellular polymeric substances (EPS) produced by microorganisms, on a surface due to either deposition and/or growth. Biofouling is defined to occur when the biofilm passes a threshold of interference negatively affecting the filtration process [7]. The development and growth occur due to the continuous availability of nutrients flowing into the system [8].

Biofouling is considered the least understood and most problematic type of fouling in affecting filtration processes despite the crescent of a significant number of studies [9].

2. Fouling characterization

Over time, the formation of fouling on the membrane surface acts as a secondary filtration layer impacting the membrane performance due to the increase of the hydraulic resistance of the system. During operation, the only information available regarding the fouling formation is represented by the decrease in performance, either a decrease in flux or an increase in pressure, without any data related to the identity of the fouling (**Figure 3**). Hence, either on a full-scale plant or in a research experiment, it is common to conduct membrane autopsy in order to analyze membrane coupons collected from the module. This approach involves two different types of analysis, (i) analytical characterization performed to identify the nature of the contaminants and (ii) visualization by means of an imaging technique to identify and quantify the fouling structure. A key aspect of fouling studies includes the analysis of the structural fouling properties [10], which can forecast the fouling layer comportment, and thus, the effect on filtration performance. The structural analysis of the fouling layer deposited on a membrane coupon consists of using imaging techniques. Most of the imaging approaches and practices to characterize the fouling morphology reported in the literature include require destructive procedures and are performed after membrane autopsy (**Figure 4**) [11, 12].

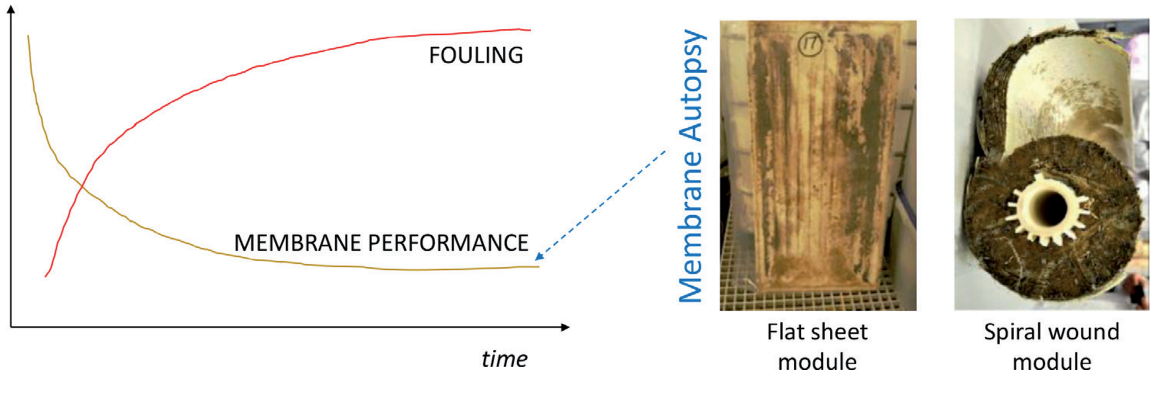

Figure 3.
Fouling development and membrane performance decrease in a membrane module.

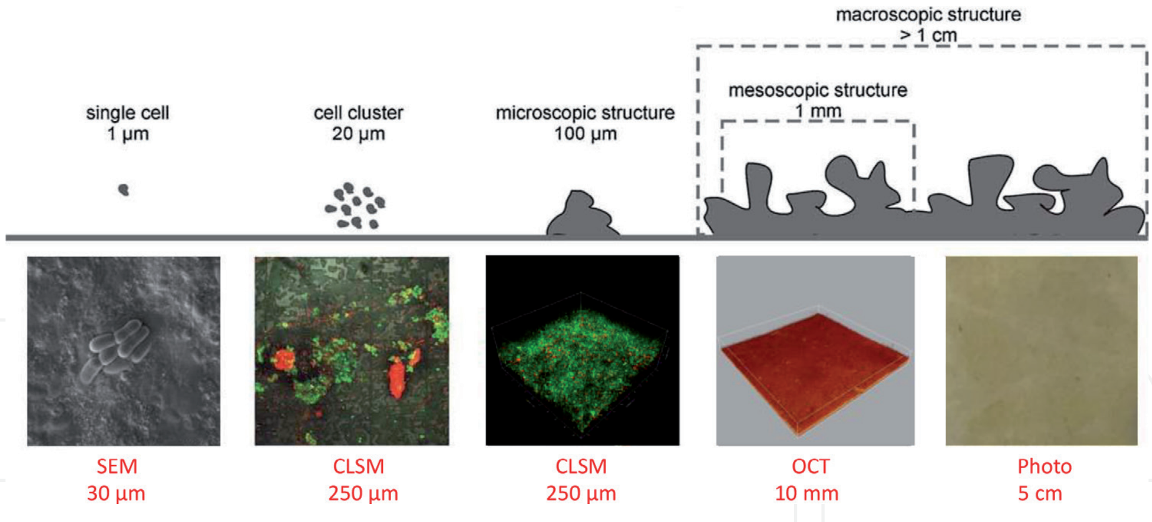

Figure 4.
Techniques to characterize fouling structure at different scales: SEM, scanning electron microscopy; CLSM, confocal laser scanning microscopy; and OCT, optical coherence tomography. Adapted from Wagner et al. [18].

Scanning electron microscopy (SEM) has been for many years the most employed technique to characterize membrane coupons. SEM is capable of visualizing the structure of fouling on a near-nanometer and sub-micron scale, allowing to distinguish among the different types of fouling, including crystals and biofilms [13, 14]. However, the SEM analysis requires the drying and coating of the analyzed sample, which represents a limitation in terms of time and artifacts. Confocal laser scanning microscopy (CLSM) has been widely used for the analysis of biological samples (biofouling). In particular, CLSM enables characterizing the constituents of biofilm, including the EPS matrix with the use of specific probes [15]. Compared to SEM, CLSM allows the three-dimensional (3D) characterization of the fouling layer [16]. However, the sample preparation for the CLSM is more complex requiring the use of specific dyes and probe, with the risk of altering and affecting the overall structure. As highlighted in the literature, all the steps involved in the staining, including storage and rinsing, can modify the biofilm structure and thus impact parameter quantification [17].

Moreover, the CLSM allows the quantification and visualization only of the stained material that is only partially representative of the fouling structure. Therefore, considering the sample handling and the staining process, the structural analysis performed by following this approach is not truly representative of fouling deposited in the system. An additional limitation is represented by the incapacity of providing information online during continuous operation.

2.1 In-situ nondestructive fouling characterization

The main limitation of the conventional imaging techniques in characterizing the fouling is represented by the impossibility of collecting data over time. In fact, SEM and CLSM are destructive analysis, requiring the destruction of the operating modules. Therefore, the destructive approach enables collecting data "only once" by destroying the membrane (**Figure 3**). Usually, the analysis is performed by membrane autopsy at the end after a significant decrease in performance, consisting in most of the cases in a decrease in permeability, that is translated in a decrease in flux under constant pressure operation and an increase in pressure under constant flux operation. Therefore, as shown in **Figure 3**, the membrane autopsy is performed usually at the end of the experiment or toward the minimum of its performances, when is "too late." In summary, conventional techniques lack in providing temporal information related to the fouling developed in the system.

Therefore, in membrane filtration processes, nondestructive in-situ biofilm fouling monitoring techniques have gained attention thanks the possibility of obtaining information regarding the fouling formed in membrane systems without stopping the process or destroying the units. Several techniques have been tested over time. The first approach consisted of employing a camera mounted on the system to monitor the membrane surface. This method allowed to evaluate the nondestructive fouling deposition and pattern at particular operating conditions. Another approach employed nuclear magnetic resonance (NMR) to detect the morphological development of biomass in membrane system [19, 20]. Recently, another approach was proposed employing planar optodes to visualize the biomass in a membrane flow cell under continuous operation by using probes. This approach enables to assess the O_2 distribution in biofilms and therefore estimate the biofouling spatial distribution [21]. Recently, the OCT has been tested as tool to evaluate the fouling deposition in membrane filtration systems in-situ nondestructively.

3. Biofilm characterization with OCT

The OCT was first developed and mainly employed in biomedical applications. The technology has been extensively used in ophthalmology for diagnosis and treatment guidance [22]. Afterward, several applications were explored, including the study of biofilm structure. In 2006, the OCT was utilized to study the growth of a *Pseudomonas aeruginosa* biofilm in a capillary flow cell [23]. Haisch and Niessner [24] evaluated the suitability of the OCT for industrial biofilm monitoring. Wagner et al. [18] showed the potentiality of a SD-OCT system OCT in characterizing the biofilm structure in the millimeters range. As highlighted in Section 1.2, the formation of biofilm is very common in membrane processes; therefore, these studies laid the foundation for studying the biomass growth in membrane processes.

Over time, the use of the OCT gained significative attention in the study of biofouling due to a series of advantages respect the other conventional techniques [11, 25]. The first benefit is due to the ability to investigate fouling formation in-situ without any staining. Indeed, the OCT is a label-free technique that enables acquiring 3D data eliminating the all risks related to the use of specific probe and chemicals [26, 27]. Another advantage consists in the possibility of acquiring information at the mesoscale level, allowing to monitor the biomass in the millimeters range. The mesoscale is considered ideal for studying the bulk-fouling interface in order

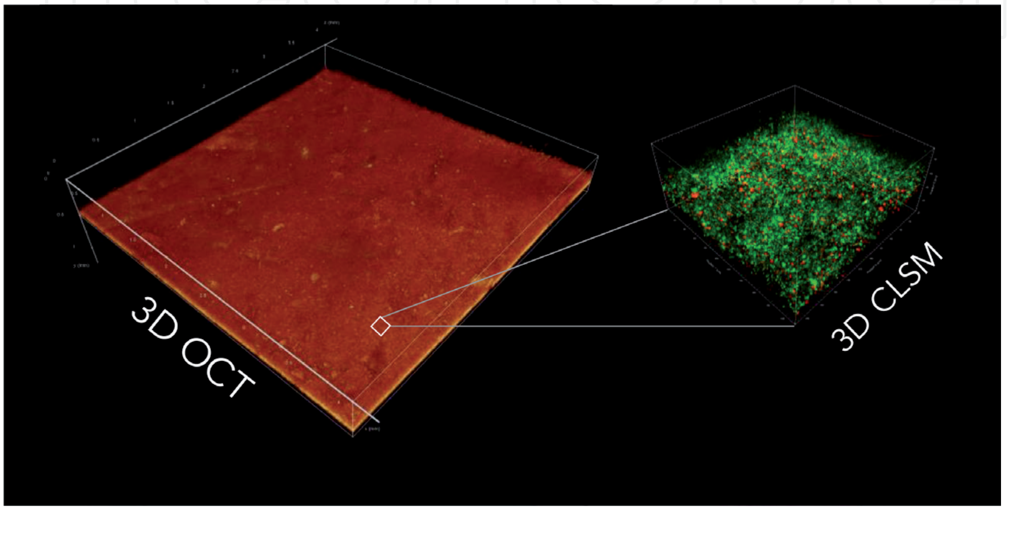

Figure 5.
3D rendered fouling structure obtained from the OCT and CLSM datasets. Adapted from Fortunato et al. [28].

to understand the fluid-structure interaction (**Figure 3**). On the other side, SEM and CLSM allow to acquire information at the microscopic level in the micrometers range. The diminution of the monitored portion increases the risk of analyzing an area not representative of the process. By using CLSM, we need to acquire more than 400 scans to cover the same area covered by a single OCT scan (**Figure 5**) [18, 28]. Moreover, it is worth to mention that the OCT does not require any time for sampling and a 3D scan can be acquired in the range of 1–2 minutes. All these features are necessary to acquire information regarding the fouling formation to assess the amount of biomass deposited in the system and its hydraulic resistance of the layer deposited on the membrane. Indeed, the main objective of the fouling characterization is to understand the impact of the biomass deposited on the system performance [10].

3.1 Morphology analysis

Besides the visualization, the main objective of the OCT in-situ observation is related to the possibility of describing and quantifying the monitored structure. A complete list of the key parameters that can be obtained from tomography dataset was previously presented by Beyenal et al. [29]. The OCT device used is in most of the studies is the Thorlabs GANYMEDE spectral domain OCT system with a central wavelength of 930 (Thorlabs, GmbH, Dachau, Germany) equipped with a 5× telecentric scan lens (Thorlabs LSM 03BB). In the case of fouling characterization, the most used parameters are: mean thickness, relative roughness, absolute roughness, membrane coverage, biovolume, and macro-porosity. The relative and absolute roughnesses are parameters used to assess the heterogeneity of the biomass morphology. The first step of the image analysis necessary to extract the data is binarization, which allows to identify and distinguish the biomass signal to the background noise. This step in the case of OCT datasets is complex since in most of the cases it is not possible to perform the segmentation based only on a pixel intensity threshold. Indeed, the polymeric support below the membrane has often the same intensity of the biomass deposited on the membrane surface. Therefore, often the intensity threshold needs to be coupled with an edge detection algorithm that allows to identify biomass, only the pixels above the membrane. Another obstacle is due to the speckle noise that hinders the biomass binarization. Frequently *mean* and *median* filters are applied before the thresholding to improve the binarization processes. However, these filters are not suitable for interferometric images and might alter the OCT dataset. Several efforts have been made to develop algorithms to denoise the OCT scans [30]. Recently, a specific algorithm was developed and successfully tested for the study and the quantification of the cake layer development in activated sludge membrane bioreactor treating real wastewater [28, 31].

An alternative strategy to simplify the binarization process and the image analysis consists in subtracting the initial OCT scan acquired at time 0 to all the other scans, in this way all the extern signals to the biomass are eliminated from the scans. This approach has been successfully employed in different cases and membrane processes [32–35].

The first parameters usually calculated from the OCT datasets is the fouling mean thickness. This is generally defined as the distance between the upper and the lower layer of the fouling interface. The value is usually expressed in pixel and then converted in μm taking into account the refractive index of the penetrated media [26]. In literature, it is assumed a single refractive index equal to 1.333. The other key descriptors extracted from the image analysis are related to the fouling homogeneity. Relative and absolute roughnesses are considered as roughness measurements where the first one is dimensionless and the second one is in μm [26, 36]. The macroporosity was also introduced as parameters to quantify the presence of big voids inside the fouling structure [36]. Another important value calculated from the analysis is the membrane coverage that expresses the amount of membrane covered by fouling [37].

3.2 Monitoring the fouling growth under continuous operation

Due to its ability to monitor the system without stopping or affecting in any ways, OCT process has been used so far in several membrane filtrations systems and configurations. The central advantage respect to the other monitoring techniques is represented by the possibility of "having a window on the system" (**Figure 6**), and thus being able to correlate the decrease in performances to the in-situ investigation. Among the different types of membrane configurations, the flat sheet membrane bioreactor is one of the easiest systems on which the OCT is employed. Flat sheet membranes are easy to install and compared to the hollow fiber, they have the advantage of being flat, therefore, keeping a fixed distance from the membrane to the OCT probe. This enables covering bigger areas on a single scan moving easily among different positions in the membrane module. The research in submerged system representative of reactors is performed on tank of Plexiglas, where the membrane is submerged. OCT was employed to evaluate the change of morphology during 42 days in a gravity-driven membrane bioreactor treating synthetic wastewater [36]. As shown in **Figure 7**, a significative change in morphology was observed over time, highlighting the evolution and the change of the fouling layer during the operation.

The possibility of capturing a fouling morphology change has been exploited by several authors. Some tests were focused on evaluating the change of morphologies linked to the change of the feed [38, 39]. Experiments are often performed in membrane flowcells and compared to a control. Shao et al. [40] compared the backwashing efficiency in controlling the biofouling by using two different backwash feeds. Derlon et al. [26] used the OCT to evaluate the effect of metazoan as biological control of biofilm developed on the membrane. The use of metazoan led to a change in fouling morphology that was also linked to an enhancement in the flux. Farid et al. [41] used the OCT to evaluate the bacterial inactivation of a graphene oxide

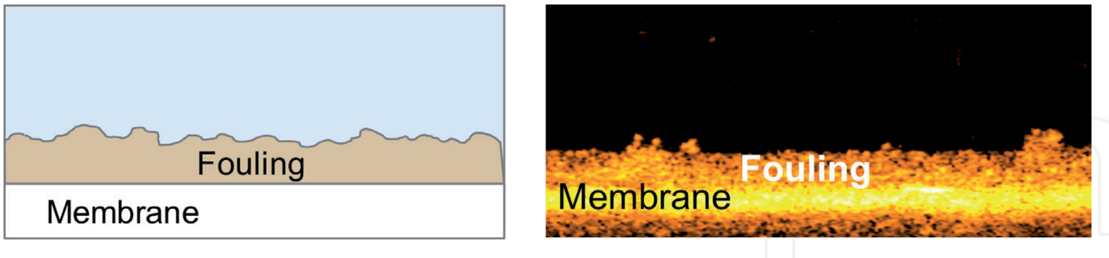

Figure 6.
Schematic representation of OCT in-situ monitoring in a membrane filtration process. 2D OCT scan acquired under continuous operation of the biomass deposited on the membrane surface.

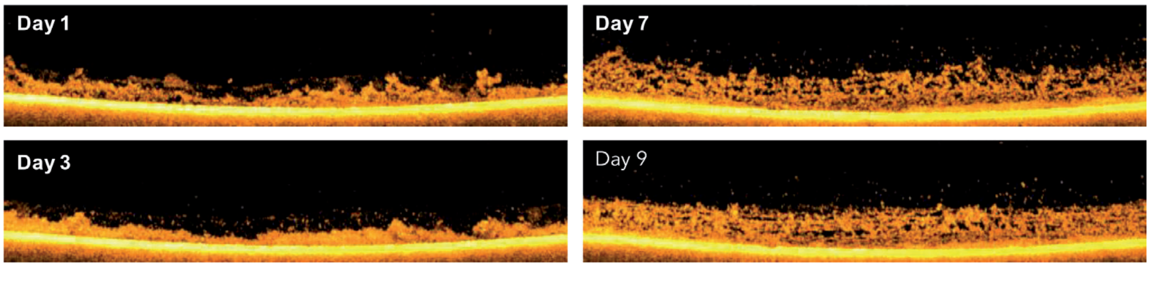

Figure 7.
Time-resolved analysis of fouling deposited on a flat sheet membrane treating wastewater. Adapted from Fortunato et al. [34].

membrane. In this case, the use of the OCT highlighted the difference in deposition between a commercial membrane and a graphene oxide membrane with antifouling proprieties. The OCT was employed to study the compression and decompression of the structure were observed and to relate to the biofilm mechanical proprieties [42, 43]. The authors observed an increase in pressure drop and hydraulic resistance over time. Desmond et al. [44] employed the OCT to study the compression of membrane biofilms in gravity-driven ultrafiltration. OCT scans were also used by Wibisono et al. [45] to evaluate the efficacy of two-phase cleaning flow in a spiral-wound element. Recently, the OCT has also been employed to validate a fluorescence-based method for the detection of biofouling at the early stage [46].

Another interesting benefit of using OCT in filtration processes is represented by the opportunity of recording the cross section at high frequency. Blauert et al. [47] used this approach to evaluate the time-resolved deformation of a biofilm in a flow cell enabling the estimation of the mechanical proprieties. Fortunato et al. [37], by using time-resolved analysis, were able to generate a video of the biomass developed during early stage filtration on a submerged membrane. The image analysis performed on the scans showed a correlation between the biomass membrane coverage and the biomass thickness with the decrease of fouling at an early stage. Moreover, through the videos, it was possible to correlate the biomass development with the flux decrease and capture particular morphologies constituted by a double-layer structure, where the upper one was moving and the lower one was still.

3.3 OCT monitoring in spacer-filled channel

The same approach was extended to spacer-filled channel spiral wound module, which is the membrane module employed for desalination. In spiral-wound membrane, biofouling has been identified as the bottleneck [48], since it leads to decrease in performances in the full-scale operating plants due to the increase in feed channel pressure drop, permeate flux reduction, and/or salt passage increase [49]. The spacer-filled channel geometry is a characteristic of spiral wound elements, where the spacer is used to increase the turbulence and separate the membrane sheets. This configuration is considered more complex for imaging purpose due to the presence of a plastic feed spacer that complicates the image processing and morphology analysis. The research in this field is carried out by using a flow cell called membrane fouling simulator (MFS) representative of the hydrodynamics, and it has all the elements present in the module. In monitoring the fouling in spacer-filled channel, the ability of the OCT in acquiring scans in the mm range resulted even essential due to the presence of the feed spacer. In fact, by using the OCT, it was possible to monitor the biomass deposited on the feed spacer pattern representative of the hydrodynamics of the module. West et al. [50] performed image analysis on 3D OCT dataset on two different feed spacer meshes. Fortunato et al. [33] visualized and quantified the 3D structure of the biomass deposited in the system, enabling to assess the spatial distribution of the biomass in the channel, whereas the highest deposition was observed on the feed spacer. Afterward, the biofilm thickness map was proposed as a tool to quickly evaluate the biomass deposited in spacer-filled channel (**Figure 8**) [34].

3.4 Monitoring inorganic fouling

In the case of membrane filtration systems, the OCT was at the beginning employed to study the biofilm and the biofouling formation, afterward the in-situ monitoring was also extended to other types of fouling. Online monitoring was

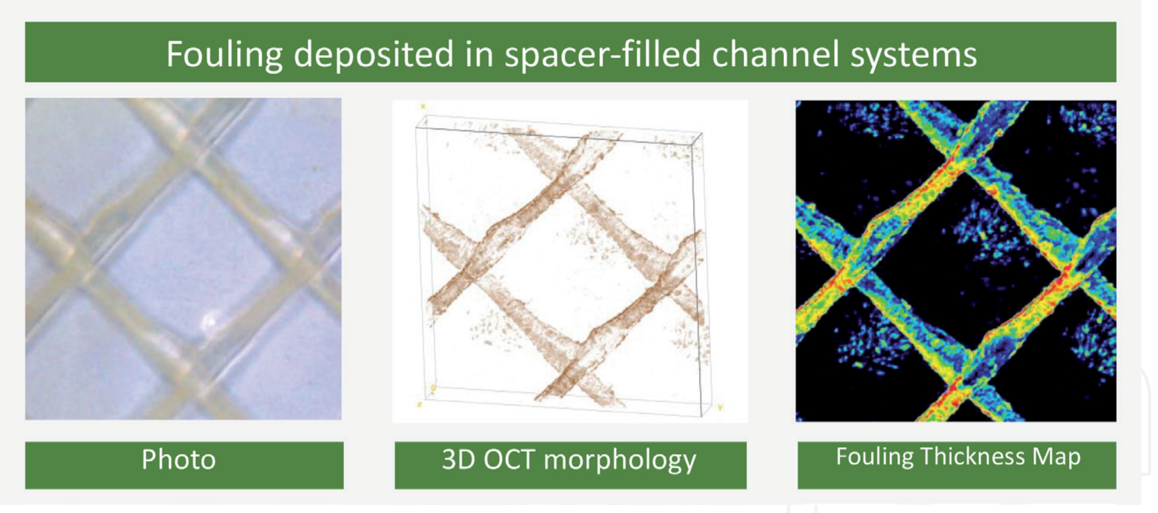

Figure 8.
Fouling characterization in spacer-filled channel systems through 3D OCT image analysis. The spacer-filled channel geometry is typical of the membrane employed for sweater desalination. Adapted from Fortunato et al. [34].

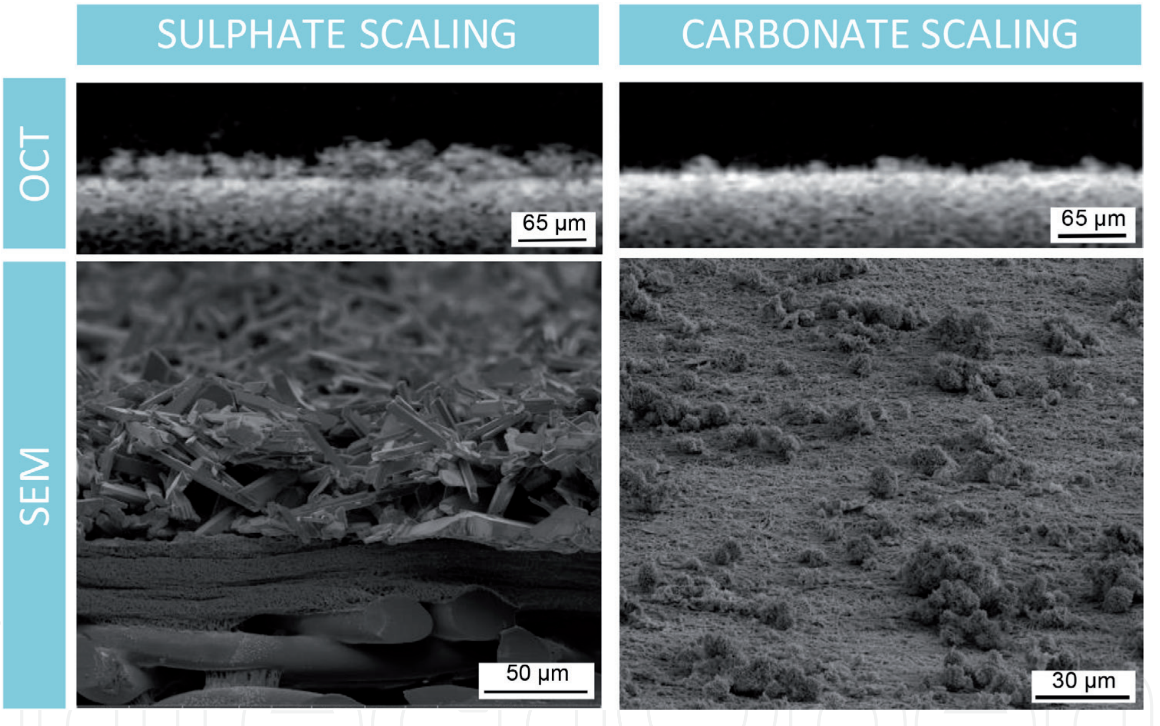

Figure 9.
Inorganic fouling structure deposited on membrane surface treating saline feed. OCT scans and SEM images. Adapted from Fortunato et al. [52].

performed on fouling layer formed by different particles as silica and bentonite [32, 51]. Recently, Fortunato et al. [52] employed the OCT to monitor the formation of scaling in a membrane distillation process. Membrane distillation is a hybrid process that couples thermal and membrane processes used to treat high saline feed as the brine. In these studies, the OCT analysis was coupled with a membrane autopsy to identify the nature of the deposition. Thought the in-situ analysis was possible to evaluate the formation of carbonate and sulfate crystals over time (**Figure 9**). The use of OCT was then proposed as a tool to monitor the scaling in thermal process that employs membrane [53]. Recently, Bauer et al. [54] used the OCT to quantify the area covered by the scaling and the flux decline in a membrane distillation process. In 2018, the OCT was employed to analyze the external and internal fouling due to oil droplets [55].

3.5 Using the OCT to improve fluid dynamic simulation

One of the main objectives of the fouling structural analysis is to evaluate the impact of the biomass deposited on the membrane on the performance. Modeling is often performed on filtration process to evaluate the effect of the shear force on the biomass formed and provide a better understanding of the process. Furthermore, there has always been a demand to implement the biomass structure in multidimensional models to understand the structure-fluid interaction and predict the behavior. Indeed, a real biomass structure better matches with the process performance respect to a theoretical structure with a given average thickness. Initially, the acquisition of the structure that affected the membrane permeability was performed by means of the CLSM [56]; however, as stated in Section 2, the CLSM has several disadvantages with respect to the OCT, including the incapability of acquiring data nondestructively.

Moreover, OCT enables to acquire information at wider scale allowing to study an area more representative of the process. The OCT scans were used by Martin et al. [57] to perform simulation of the permeate flux in a gravity driven system. Gao et al. [58] used the Doppler effect to visualize the velocity field in a spacer-filled channel. Fortunato et al. [36] imported the real biomass morphologies developed on a membrane bioreactor-treated secondary wastewater effluent. The biomass morphology was imported after 3 and 30 days of filtration and implemented in a computational fluid dynamic simulation (CFD), allowing to identify the local region of local and high flux within the biomass structure. The approach proposed allowed to match the model with the experimental values of the permeate fluxes. Jafari et al. [59] developed a numerical model able to correlate the structural deformation with biofilm hydraulics by using the in-situ observation performed with OCT. Recently, Picioreanu et al. [60] employed the OCT scans to develop a method for the determination of the elastic proprieties of a biofilm. In summary, coupling the OCT with the CFD represents a powerful toolbox to understand and predict the behavior of the biomass in membrane filtration processes.

4. Conclusions

Membrane fouling is considered the main limitation of membrane filtration systems in terms of cost and operation. The techniques commonly used for analyzing the fouling are based on membrane autopsies, where membrane coupons were collected and analyzed after destroying a membrane module. With that approach, therefore, it is possible to provide information only at the specific time chosen to conduct the autopsy. Those techniques are consequently subject to the circumstance of ending the process. Moreover, some of the techniques necessitate sampling preparation such as drying or labeling, which have the risk of changing the fouling morphology. Therefore, considering that the fouling is a dynamic process and will evolve, it is necessary to monitor the fouling development over time under continuous operation without interfering with the process.

Thanks to the possibility of monitoring samples without the use of staining in-situ nondestructively, the use of OCT gained attention in studying the fouling in membrane filtration systems. At the beginning, the OCT was employed to study the biofilm formed in the process and later applied to all the different types of fouling. Nowadays, the OCT is considered an essential tool to gain a better understanding of fouling behavior and is employed in different membrane configurations and systems covering the whole spectrum of membrane filtration processes. The in-situ nondestructive online acquisition cross-sectional scans of the fouling deposited enables to link the impact of the fouling on the membrane performance (i.e., flux decrease and feed channel pressure drop). The approach also resulted to be beneficial in evaluating the efficacy of antifouling strategies. In summary, the use of OCT in membrane filtration systems turned out to be a key tool in understanding and predicting the fouling development and its effect on the overall membrane performance.

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Conflict of interest

I confirm there are no conflicts of interest.

Author details

Luca Fortunato
Division of Biological and Environmental Science and Engineering (BESE), Water Desalination and Reuse Center (WDRC), King Abdullah University of Science and Technology (KAUST), Thuwal, Saudi Arabia

*Address all correspondence to: luca.fortunato@kaust.edu.sa

References

[1] Rost S, Gerten D, Bondeau A, Lucht W, Rohwer J, Schaphoff S. Agricultural green and blue water consumption and its influence on the global water system. Water Resources Research. 2008;**44**. DOI: 10.1029/2007WR006331

[2] U.& WWAP. The United Nations World Water Development Report 2;2006

[3] Baker RW. Membrane Technology and Applications2012. DOI: 10.1002/9781118359686

[4] Drews A. Membrane fouling in membrane bioreactors—Characterisation, contradictions, cause and cures. Journal of Membrane Science. 2010;**363**:1-28. DOI: 10.1016/j.memsci.2010.06.046

[5] Le-Clech P, Chen V, Fane TAG. Fouling in membrane bioreactors used in wastewater treatment. Journal of Membrane Science. 2006;**284**:17-53. DOI: 10.1016/j.memsci.2006.08.019

[6] Nguyen T, Roddick FA, Fan L. Biofouling of water treatment membranes: A review of the underlying causes, monitoring techniques and control measures. Membranes (Basel). 2012;**2**:804-840. DOI: 10.3390/membranes2040804

[7] Flemming HC. Biofouling in water systems—Cases, causes and countermeasures. Applied Microbiology and Biotechnology. 2002;**59**:629-640. DOI: 10.1007/s00253-002-1066-9

[8] Flemming HC, Schaule G, Griebe T, Schmitt J, Tamachkiarowa A. Biofouling—The Achilles heel of membrane processes. Desalination. 1997;**113**:215-225. DOI: 10.1016/S0011-9164(97)00132-X

[9] Weinrich L, Haas CN, LeChevallier MW. Recent advances in measuring and modeling reverse osmosis membrane fouling in seawater desalination: A review. Journal of Water Reuse and Desalination. 2013;**3**:85. DOI: 10.2166/wrd.2013.056

[10] Halan B, Buehler K, Schmid A. Biofilms as living catalysts in continuous chemical syntheses. Trends in Biotechnology. 2012;**30**:453-465. DOI: 10.1016/j.tibtech.2012.05.003

[11] Valladares Linares R, Fortunato L, Farhat NM, Bucs SS, Staal M, Fridjonsson EO, et al. Mini-review: Novel non-destructive in situ biofilm characterization techniques in membrane systems. Desalination and Water Treatment. 2016;**57**:22894-22901. DOI: 10.1080/19443994.2016.1180483

[12] Fortunato L, Jeong S, Wang Y, Behzad AR, Leiknes T. Integrated approach to characterize fouling on a flat sheet membrane gravity driven submerged membrane bioreactor. Bioresource Technology. 2016;**222**:335-343. DOI: 10.1016/j.biortech.2016.09.127

[13] Lawrence JR, Swerhone GDW, Leppard GG, Araki T, Zhang X, West MM, et al. Scanning transmission X-ray, laser scanning, and transmission electron microscopy mapping of the exopolymeric matrix of microbial biofilms. Applied and Environmental Microbiology. 2003;**69**:5543-5554. DOI: 10.1128/AEM.69.9.5543-5554.2003

[14] Herzberg M, Elimelech M. Biofouling of reverse osmosis membranes: Role of biofilm-enhanced osmotic pressure. Journal of Membrane Science. 2007;**295**:11-20. DOI: 10.1016/j.memsci.2007.02.024

[15] Flemming HC, Wingender J. The biofilm matrix. Nature Reviews. Microbiology. 2010;**8**:623-633. DOI: 10.1080/0892701031000072190

[16] Lewandowski Z, Webb D, Hamilton M, Harkin G. Quantifying biofilm structure. Water Science and Technology. 1999;**39**:71-76. DOI: 10.1016/S0273-1223(99)00152-3

[17] Jin X, Shan J, Wang C, Wei J, Tang CY. Rejection of pharmaceuticals by forward osmosis membranes. Journal of Hazardous Materials. 2012;**227-228**:55-61. DOI: 10.1016/j.jhazmat.2012.04.077

[18] Wagner M, Taherzadeh D, Haisch C, Horn H. Investigation of the mesoscale structure and volumetric features of biofilms using optical coherence tomography. Biotechnology and Bioengineering. 2010;**107**:844-853. DOI: 10.1002/bit.22864

[19] Graf von der Schulenburg DA, Vrouwenvelder JS, Creber SA, van Loosdrecht MCM, Johns ML. Nuclear magnetic resonance microscopy studies of membrane biofouling. Journal of Membrane Science. 2008;**323**:37-44. DOI: 10.1016/j.memsci.2008.06.012

[20] Fridjonsson EO, Vogt SJ, Vrouwenvelder JS, Johns ML. Early non-destructive biofouling detection in spiral wound RO membranes using a mobile earth's field NMR. Journal of Membrane Science. 2015;**489**:227-236. DOI: 10.1016/j.memsci.2015.03.088

[21] Frederiksen MS, Glud RN. Oxygen dynamics in the rhizosphere of Zostera marina: A two-dimensional planar optode study. Limnology and Oceanography. 2006;**51**:1072-1083

[22] Wojtkowski M, Srinivasan V, Fujimoto JG, Ko T, Schuman JS, Kowalczyk A, et al. Three-dimensional retinal imaging with high-speed ultrahigh-resolution optical coherence tomography. Ophthalmology. 2005;**112**:1734-1746. DOI: 10.1016/j.ophtha.2005.05.023

[23] Xi C, Marks D, Schlachter S, Luo W, Boppart S a. High-resolution three-dimensional imaging of biofilm development using optical coherence tomography. Journal of Biomedical Optics. 2006;**11**:034001. DOI: 10.1117/1.2209962

[24] Haisch C, Niessner R. Visualisation of transient processes in biofilms by optical coherence tomography. Water Research. 2007;**41**:2467-2472. DOI: 10.1016/j.watres.2007.03.017

[25] Wagner M, Horn H. Optical coherence tomography in biofilm research: A comprehensive review. Biotechnology and Bioengineering. 2017;**114**:1386-1402. DOI: 10.1002/bit.26283

[26] Derlon N, Peter-Varbanets M, Scheidegger A, Pronk W, Morgenroth E. Predation influences the structure of biofilm developed on ultrafiltration membranes. Water Research. 2012;**46**:3323-3333. DOI: 10.1016/j.watres.2012.03.031

[27] Dreszer C, Flemming HC, Zwijnenburg A, Kruithof JC, Vrouwenvelder JS. Impact of biofilm accumulation on transmembrane and feed channel pressure drop: Effects of crossflow velocity, feed spacer and biodegradable nutrient. Water Research. 2014;**50**:200-211. DOI: 10.1016/j.watres.2013.11.024

[28] Fortunato L, Li M, Cheng T, Rehman ZU, Heidrich W, Leiknes T. Cake layer characterization in activated sludge membrane bioreactors: Real-time analysis. Journal of Membrane Science. 2019;**578**:163-171. DOI: 10.1016/J.MEMSCI.2019.02.026

[29] BeyenalH,DonovanC,LewandowskiZ, Harkin G. Three-dimensional biofilm structure quantification. Journal of Microbiological Methods. 2004;**59**:395-413. DOI: 10.1016/J.MIMET.2004.08.003

[30] Li M, Idoughi R, Choudhury B, Heidrich W. Statistical model for OCT

image denoising. Biomedical Optics Express. 2017;**8**:3903. DOI: 10.1364/BOE.8.003903

[31] Fortunato L, Pathak N, Ur Rehman Z, Shon H, Leiknes T. Real-time monitoring of membrane fouling development during early stages of activated sludge membrane bioreactor operation. Process Safety and Environment Protection. 2018;**120**:313-320. DOI: 10.1016/J.PSEP.2018.09.022

[32] Gao Y, Haavisto S, Li W, Tang CY, Salmela J, Fane AG. Novel approach to characterizing the growth of a fouling layer during membrane filtration via optical coherence tomography. Environmental Science and Technology. 2014;**48**:14273-14281. DOI: 10.1021/es503326y

[33] Fortunato L, Bucs S, Linares RV, Cali C, Vrouwenvelder JS, Leiknes T. Spatially-resolved in-situ quantification of biofouling using optical coherence tomography (OCT) and 3D image analysis in a spacer filled channel. Journal of Membrane Science. 2017;**524**:673-681. DOI: 10.1016/j.memsci.2016.11.052

[34] Fortunato L, Leiknes TO. In-situ biofouling assessment in spacer filled channels using optical coherence tomography (OCT): 3D biofilm thickness mapping. Bioresource Technology. 2017;**229**:231-235. DOI: 10.1016/j.biortech.2017.01.021

[35] Li W, Liu X, Wang Y-N, Chong TH, Tang CY, Fane AG. Analyzing the evolution of membrane fouling via a novel method based on 3D optical coherence tomography imaging. Environmental Science and Technology. 2016;**50**:6930-6939. DOI: 10.1021/acs.est.6b00418

[36] Fortunato L, Qamar A, Wang Y, Jeong S, Leiknes T. In-situ assessment of biofilm formation in submerged membrane system using optical coherence tomography and computational fluid dynamics. Journal of Membrane Science. 2017;**521**:84-94. DOI: 10.1016/j.memsci.2016.09.004

[37] Fortunato L, Jeong S, Leiknes T. Time-resolved monitoring of biofouling development on a flat sheet membrane using optical coherence tomography. Scientific Reports. 2017;**7**:15. DOI: 10.1038/s41598-017-00051-9

[38] Pathak N, Fortunato L, Li S, Chekli L, Phuntsho S, Ghaffour N, et al. Evaluating the effect of different draw solutes in a baffled osmotic membrane bioreactor-microfiltration using optical coherence tomography with real wastewater. Bioresource Technology. 2018;**263**:306-316. DOI: 10.1016/j.biortech.2018.04.123

[39] Wang Y, Fortunato L, Jeong S, Leiknes TO. Gravity-driven membrane system for secondary wastewater effluent treatment: Filtration performance and fouling characterization. Separation and Purification Technology. 2017;**184**:26-33. DOI: 10.1016/j.seppur.2017.04.027

[40] Shao S, Wang Y, Shi D, Zhang X, Tang CY, Liu Z, et al. Biofouling in ultrafiltration process for drinking water treatment and its control by chlorinated-water and pure water backwashing. Science of the Total Environment. 2018;**644**:306-314. DOI: 10.1016/J.SCITOTENV.2018.06.220

[41] Farid MU, Guo J, An AK. Bacterial inactivation and in situ monitoring of biofilm development on graphene oxide membrane using optical coherence tomography. Journal of Membrane Science. 2018;**564**:22-34. DOI: 10.1016/J.MEMSCI.2018.06.061

[42] Valladares Linares R, Wexler AD, Bucs SS, Dreszer C, Zwijnenburg A, Flemming HC, et al. Compaction and relaxation of biofilms. Desalination and Water Treatment. 2016;**57**:12902-12914. DOI: 10.1080/19443994.2015.1057036

[43] Dreszer C, Wexler AD, Drusová S, Overdijk T, Zwijnenburg A, Flemming HC, et al. In-situ biofilm characterization in membrane systems using optical coherence tomography: Formation, structure, detachment and impact of flux change. Water Research. 2014;**67**:243-254. DOI: 10.1016/j.watres.2014.09.006

[44] Desmond P, Morgenroth E, Derlon N. Physical structure determines compression of membrane biofilms during gravity driven membrane (GDM) ultrafiltration. Water Research. 2018;**143**:539-549. DOI: 10.1016/J.WATRES.2018.07.008

[45] Wibisono Y, El Obied KE, Cornelissen ER, Kemperman AJB, Nijmeijer K. Biofouling removal in spiral-wound nanofiltration elements using two-phase flow cleaning. Journal of Membrane Science. 2015;**475**:131-146. DOI: 10.1016/j.memsci.2014.10.016

[46] Khan BK, Fortunato L, Leiknes T. Early biofouling detection using fluorescence-based extracellular enzyme activity. Enzyme and Microbial Technology. 2019;**120**:43-51. DOI: 10.1016/J.ENZMICTEC.2018.10.001

[47] Blauert F, Horn H, Wagner M. Time-resolved biofilm deformation measurements using optical coherence tomography. Biotechnology and Bioengineering. 2015;**112**:1893-1905. DOI: 10.1002/bit.25590

[48] Vrouwenvelder JS, Bakker SM, Wessels LP, van Paassen JAM. The membrane fouling simulator as a new tool for biofouling control of spiral-wound membranes. Desalination. 2007;**204**:170-174. DOI: 10.1016/j.desal.2006.04.028

[49] Characklis W, Marshall K. Biofilms. New York: John Wiley & Sons; 1990

[50] West S, Wagner M, Engelke C, Horn H. Optical coherence tomography for the in situ three-dimensional visualization and quantification of feed spacer channel fouling in reverse osmosis membrane modules. Journal of Membrane Science. 2015;**498**:345-352. DOI: 10.1016/j.memsci.2015.09.047

[51] Li C, Wagner M, Lackner S, Horn H. Assessing the influence of biofilm surface roughness on mass transfer by combining optical coherence tomography and two-dimensional modeling. 2016;**113**:989-1000. DOI: 10.1002/bit.25868

[52] Fortunato L, Jang Y, Lee JG, Jeong S, Lee S, Leiknes TO, et al. Fouling development in direct contact membrane distillation: Non-invasive monitoring and destructive analysis. Water Research. 2018;**132**:34-41. DOI: 10.1016/j.watres.2017.12.059

[53] Lee JG, Jang Y, Fortunato L, Jeong S, Lee S, Leiknes T, et al. An advanced online monitoring approach to study the scaling behavior in direct contact membrane distillation. Journal of Membrane Science. 2018;**546**:50-60. DOI: 10.1016/J.MEMSCI.2017.10.009

[54] Bauer A, Wagner M, Saravia F, Bartl S, Hilgenfeldt V, Horn H. In-situ monitoring and quantification of fouling development in membrane distillation by means of optical coherence tomography. Journal of Membrane Science. 2019;**577**:145-152. DOI: 10.1016/J.MEMSCI.2019.02.006

[55] Trinh TA, Li W, Han Q, Liu X, Fane AG, Chew JW. Analyzing external and internal membrane fouling by oil emulsions via 3D optical coherence tomography. Journal of Membrane Science. 2018;**548**:632-640. DOI: 10.1016/J.MEMSCI.2017.10.043

[56] Horn H, Lackner S. Modeling of biofilm systems: A review. In: Muffler K, Ulber R, editors. Productive Biofilms. Cham: Springer International Publishing; 2014. pp. 53-76. DOI: 10.1007/10_2014_275

[57] Martin KJ, Bolster D, Derlon N, Morgenroth E, Nerenberg R. Effect of fouling layer spatial distribution on permeate flux: A theoretical and experimental study. Journal of Membrane Science. 2014;**471**:130-137. DOI: 10.1016/j.memsci.2014.07.045

[58] Gao Y, Haavisto S, Tang CY, Salmela J, Li W. Characterization of fluid dynamics in spacer-filled channels for membrane filtration using Doppler optical coherence tomography. Journal of Membrane Science. 2013;**448**:198-208. DOI: 10.1016/j.memsci.2013.08.011

[59] Jafari M, Desmond P, van Loosdrecht MCM, Derlon N, Morgenroth E, Picioreanu C. Effect of biofilm structural deformation on hydraulic resistance during ultrafiltration: A numerical and experimental study. Water Research. 2018;**145**:375-387. DOI: 10.1016/J.WATRES.2018.08.036

[60] Picioreanu C, Blauert F, Horn H, Wagner M. Determination of mechanical properties of biofilms by modelling the deformation measured using optical coherence tomography. Water Research. 2018;**145**:588-598. DOI: 10.1016/J.WATRES.2018.08.070