

# Recycled-Membrane Biofilm Reactors based on end-of-life reverse osmosis membranes as a novel low-cost alternative for microcystins removal.

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## Introduction

The cyanobacterial proliferation as harmful blooms is becoming increasingly common issue as a consequence of climate change and freshwater eutrophication. One of the most important troublesome is the toxin production by certain cyanobacterial species. Specially, the production of Microcystins (MCs), a highly toxic and worldwide spread cyanotoxin, which cause hepatic damage and tumor promotion in animals, humans beings included. Conventional water treatments have been found inadequate for its correct and optimal elimination. For these reason, authorities around the world recommend the use of auxiliary treatments, among which have been appeared systems based on biological elimination (Chorus and Bartram, 1999).

Membrane Biofilm Reactors (MBfR) is an emerging technology based on membranes, which offers a high-performance biological alternative to remove pollutants from water. MBfR employs bacterial biofilm immobilized onto gas-permeable membranes for this purpose. In this manner, the biological activity is stimulated by supplying electron acceptor or donors across the membrane, achieving high pollutant degradation efficiencies (Martin et al., 2012). MBfRs could work with different types of membranes, such as dense, porous and composite membranes. Composite ones are the most effective due to their minor gas resistance together with a bubbleless aeration, without clogging and wetting. However, these membranes are more expensive than the others in terms of manufacturing.

At global scale, there is a huge amount of desalination RO membranes discarded every year, which fastest current solution is landfilling or incineration (Landaburu-Aguirre et al., 2016). An alternative solution to solve this growing environmental problem could be the use of discarded RO membranes for attaching MC-degrading bacteria and carry out the pollutant elimination in a novel system concept "Recycled-Membrane Biofilm Reactor" (R-MBfR) (Morón-López et al., 2019). This system could be economically competitive with other physic-chemical processes for MCs removal.

The aim of the present work is to demonstrate that MC-degrading biofilms attached onto recycled polyamide thin-film composite membranes could be stimulated by air diffusion and works like-MBfRs. So we provide new knowledge for optimizing our R-MBfR concept, offering an alternative solution for cyanotoxins removal and giving to all discarded desalination membranes modules a second life under the circular economy principles.

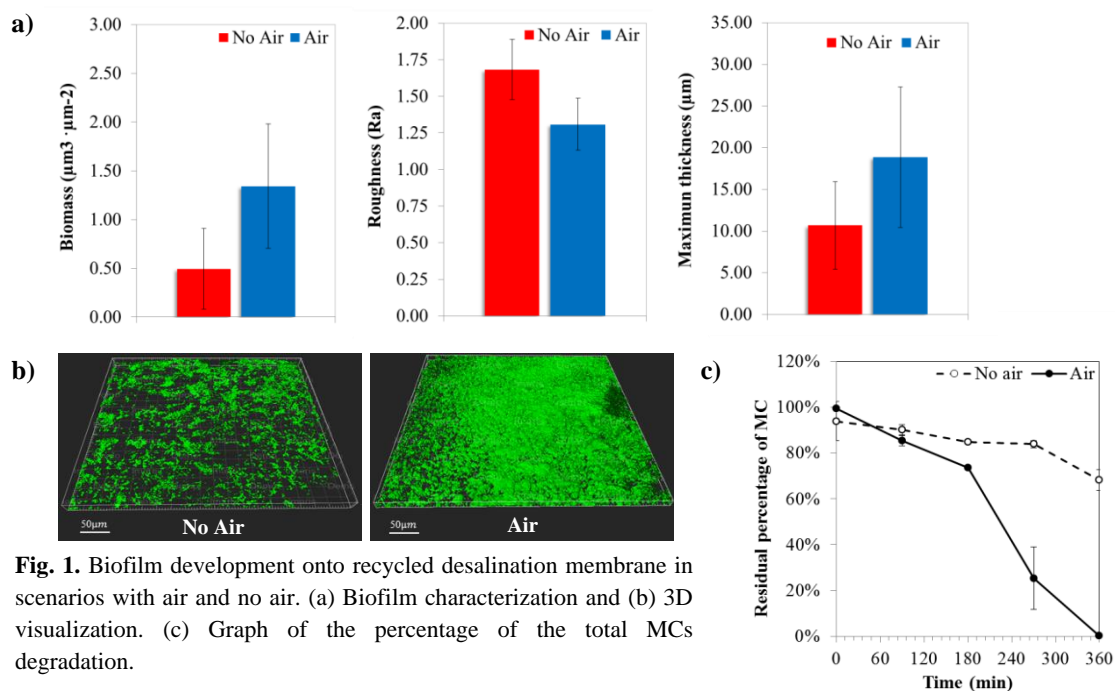
## Material and methods

Experiments were performed using EoL RO membranes originally used for seawater desalination (model HSWC3 from Hydranautics). This composite membrane has a configuration of three layers: a non-woven polyester support, an asymmetric porous polysulfone interlayer and a dense polyamide ultrathin top layer. The MC-degrading bacterial strain selected for this study was *Sphingopyxis* sp. strain IM-1, which has already been demonstrated its high capacity for MCs removal. The bacterial deposition test were carried out at laboratory scale in a simulator cell (R-MBfR simulator). The experimental setup includes a continuous feeding with R2A medium enriched with 0.01 M CaCl<sub>2</sub> at a rate of 1.4 mL·min<sup>-1</sup>, at room temperature and air diffusion from the polyester support layer (when was required). After 96 hours of growing, different samples of membranes were taken for microscopy visualization and MC degradation test. Biofilms were observed under a confocal laser microscope (CLSM, Leica SP5, Leica Microsystems) using the Live/dead BacLight Bacterial Viability Kit (Molecular Probes™) according to our previous study (Morón-López et al., 2019). On the other hand, a MC degradation test was run in a minimal salt medium (MSM) with approximately 400 mg·L<sup>-1</sup> of MCs mixture (sum of variants -LR, -RR and -YR) and following all steps also previously reported. The MCs quantification were performed using a HPLC-MS-TOF (Agilent 6230 accurate mass TOF Agilent Technology, Santa Clara, CA, USA).

## Result and discussion

The air permeability of the tested recycled composite membrane was 4.89·10<sup>-9</sup> Barrers. Accordingly, our results show that this membrane allow competitive air flux in comparison to other composite

membranes especially made for gas permeation from literature (Cerqueira et al., 2013; Motlagh et al., 2008). Besides, this air diffusion stimulated the bacterial attachment onto the membrane, as show the Fig. 1 (a) and (b). The average of the observed biomass was near to 3-fold higher when air was applied, while the average of the achieved maximum thickness was about 2-folds higher. Similarly, roughness coefficient showed more homogeneous bacterial coverage with air diffusion, maybe as a result of a more advanced biofilm stage achieved. It is also remarkable that more advanced biofilms with more thickness could be achieved in longer-time experiment, but we have focused only in early biofilms stages where the membrane surface has an important role. On the other hand, our results also showed that an enhancement of MC removal was obtained by diffusing air. The Fig.1 (c) shows that the MC degradation test using air removed the 100% of the MC concentration after 360 minutes, while only the 20% of the total MC was removed without applying air.



**Fig. 1.** Biofilm development onto recycled desalination membrane in scenarios with air and no air. (a) Biofilm characterization and (b) 3D visualization. (c) Graph of the percentage of the total MCs degradation.

## Conclusion

As far as our laboratory scale results show, the air diffusion into R-MBfR simulator may provide a high-performance system for MCs removal. The use of EoL membranes in these reactors could offer a low-cost solution, turning a waste into a resource and taking advantage of the optimal conditions offered by composite membranes in MBfRs. Therefore, it could be concluded that the R-MBfR could be a promising option for biological auxiliary treatment when needed in water treatments plants.

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