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Permeate recirculation impact on concentration polarization and fouling on RO purification of olive mill wastewater

J.M. Ochando-Pulido [a,*,1], G. Hodaifa [b], A. Martínez-Ferez [a,1]

[a] Chemical Engineering Department, University of Granada, 18071 Granada, Spain

[b] Molecular Biology and Biochemical Engineering Department, University Pablo Olavide, 14013 Seville, Spain

HIGHLIGHTS

- Impacts of permeate recirculation on membrane polarization and fouling were studied.
- Permeate recirculation reduces polarization and fouling build-up on both membranes.
- Severe fouling on low-pressure membrane stabilized upon 30% permeate recirculation.
- Irreversible fouling minimized permitting satisfactory restoration of permeability.
- 15.8–42.1% fouling build-up reduction for SC membrane whereas 22.9–55.2% for AK

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ABSTRACT

Analysis of permeate recirculation impact on membrane concentration polarization and fouling was studied upon well-controlled reverse osmosis (RO) experiments for final purification of secondary-treated olive mill wastewater (OMW). Two commercial RO membranes presenting different characteristics, that is, a thin-film composite (TFC) and a low-pressure one, were selected for comparison purposes. Incrementing the permeate recirculation ratio to the feed tank (10–30%) reduced the concentration polarization and fouling build-up, that is 15.8–42.1% for the TFC membrane whereas 22.9–55.2% for the low-pressure membrane. This fact is especially relevant for the low-pressure membrane on which fouling triggers rapidly causing severe flux loss, but tends to stabilize upon 30% permeate recirculation. Moreover, irreversible fouling was satisfactorily minimized, reducing scaling formation and permitting restoration of the initial permeability. The fouling index can be minimized by 57.1% for the TFC membrane by recirculating 30% of the permeate stream (measured at 15 °C), and also 47.1% reduction of the fouling index was observed for the low-pressure membrane upon the same recirculation ratio. The data obtained in this study highlight permeate recirculation as an important operating variable for membrane fouling control in the final RO purification of OMW.

1. Introduction

Nowadays, pressure-driven membrane processes are widely applied in water purification and industrial wastewater reclamation treatment [1–25]. Nevertheless, in spite of the high efficiency and minor investment and maintenance costs of membrane processes, concentration polarization and fouling phenomena, closely related to each other, remain still today as the main hindrance to the definite development of this broadly applied technology. This especially concerns liquid–liquid separation processes as in the case of wastewater treatment [11–13].

Microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) have already been applied for the management of the effluents derived from industries of several sectors, e.g., stainless steel [6], energy cogeneration [7], nuclear-power [8], oily wastewater [9], coking wastewater [10,11], pharmaceutical [12], textile and tannery [13–17] pulp and paper [18–20] and agro-food industries such as tomato [21], olive oil [22–25], beverage [26] and dairy production [27,28], among others.

During operation, the increase in the concentration of pollutants within the membrane boundary region leads to concentration polarization on one hand, which causes additional resistance and thus increases the operating costs and also adversely affects produced water quality. On the other hand, membrane fouling causes long-term permeate flux decline. Fouling is a complex phenomenon which may involve membrane pore plugging and clogging, chemical degradation and cake formation on the membrane surface caused by microorganisms as well as organic and inorganic material [29,30]. Fouling leads not only to an increase in the energy costs to maintain the target permeate production, but also in the operating expenses due to the need of

* Corresponding author.
E-mail address: jmochandop@ugr.es (J.M. Ochando-Pulido).
[1] Tel.: +34 958241581; fax: +34 958248992.

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frequent plant shut-downs for in-situ membrane cleaning. The longevity of the membranes can also be irretrievably shortened due to irreversible fouling.

Furthermore, to overcome the loss of performance due to fouling build-up and ensure the target steady-state permeate production, engineers erroneously tend to overdesign excessively the membrane plants, resulting in sensible but useless increment of total costs. On the other hand, under-design due to underestimation of the fouling issues leads to operation above threshold conditions, which are technically and economically unfeasible for long periods of time [31,32].

Several factors have been recognized to determine concentration polarization and fouling issues, such as the membrane type, the membrane surface roughness and mean porosity, the hydrodynamic conditions and the effluent composition and concentration [33–35].

Uncontrolled disposal of olive mill wastewater (OMW) is becoming a serious environmental hazard for an increasing number of regions, leading to problems in relation to odor nuisance, soil contamination, underground leakage and water body pollution.

The high organic pollutant concentration present in OMW, including phenols, organic acids, tannins and organohalogenated contaminants, makes these effluents phytotoxic and thus recalcitrant to biological degradation [36–40]. Moreover, OMW also exhibits high electroconductivity (EC) owed to the high concentration of inorganic material: chloride, sulfate and phosphoric salts of potassium, calcium, iron, magnesium, sodium, copper and traces of other elements [22,23,25].

Other treatment practices have been developed in time, such as lagooning or natural evaporation and thermal concentration [41,42], treatments with lime and clay [43,44], composting [45–47], biosorption [48,49], physico-chemical procedures such as coagulation–flocculation [50–52] and electrocoagulation [53,54], advanced oxidation processes including ozonation [55], Fenton's reagent [56,57] and photocatalysis [58] and also electrochemical [59–61] and hybrid processes [62–65].

In former work by the authors [66,67], the final purification of the effluents from olive oil factories (OMW) by polymeric RO membranes was addressed. OMW was primarily conducted to a secondary treatment comprising sequentially Fenton-like advanced oxidation, flocculation–sedimentation and olive stone filtration [48,49,51,56,57]. The secondary treatment ensured major organic matter abatement. Nevertheless, it was not able to reduce the high concentration of dissolved monovalent and divalent ions, which cannot be removed by conventional physicochemical treatments. Furthermore, higher conductivity values were noticed when compared to the raw OMW. This was mainly owed to the increment in sodium and chloride content derived from the addition of the neutralizing agent and the catalyst into the flocculation and oxidation tanks, respectively. Membrane technology, in virtue of its versatility and modular nature [68], can represent a cost-effective solution for the reclamation of OMW/ST.

In this research paper, the analysis of the impacts of permeate recirculation on membrane concentration polarization as well as reversible and irreversible fouling were studied upon well-controlled dynamic RO experiments. For this purpose, two commercial polymeric RO membranes presenting different characteristics, that is a thin-film composite (TFC) RO membrane previously used in other work by the authors [66, 67] and a low-pressure RO membrane, were selected for comparison purposes.

2. Experimental

2.1. Analytical methods

Analytical grade reagents were employed for the analytical proceedings, which were triplicated. Chemical oxygen demand (COD), total suspended solids (TSS), total phenols (TPh), total iron, electroconductivity (EC) and pH measurements were assessed following standard methods [69].

EC and pH measurements were performed with a Crison GLP31 conductivity-meter and a Crison GLP21 pH-meter, whereas a Helios Gamma UV–visible spectrophotometer (Thermo Fisher Scientific) served for COD, TPh and total iron measurements (Standard German methods ISO 8466-1 and German DIN 38402 A51) [69]. Ionic concentrations were analyzed with a Dionex DX-120 ion chromatograph, as described in previous work [67].

Microphotographs of the active layer of the virgin membranes and elemental microanalysis of the fouled membranes after the RO experiments were performed with a high resolution scanning electron (HR-SEM) microscope (Carl Zeiss SMT model).

2.2. The effluent stream

During the production of olive oil, two-phase continuous centrifugation-based olive oil factories lead to the generation of two main wastewater streams, the first one from the washing of the fruit (olives washing wastewater, OWW) and the second one from the olive oil washing (olive oil washing wastewater, OOW) during the vertical centrifugation. These effluents are commonly referred to as olive oil mill wastewater (OMW) [48,49,51,56,57]. An average-sized olive oil factory produces around 1 m^3 of OOW per ton of processed olives, which means a daily amount of more than 10 m^3 of OOW in sum to 1 m^3 of OWW per ton of washed olives.

Samples of OWW and OOW effluents were collected from several olive oil mills in the Andalusian provinces of Jaén and Granada (Spain) during winter months and rapidly analyzed in the lab and refrigerated for further research when necessary. OWW and OOW were mixed in 1:1 (v/v) proportion to stabilize the average organic matter concentration of the effluent stream (OMW) entering the treatment system and thus avoid sensible fluctuations in the COD parameter. After this, OMW was conducted to a secondary treatment on a pilot scale comprising Fenton-like advanced oxidation followed by flocculation–sedimentation and olive stone filtration. The secondary treatment is described in detail in former works by the authors [48,49,51,56,57]. The OMW effluent after secondary treatment will be hereafter referred as OMW/ST and presents the characteristics reported in Table 1.

2.3. Membrane operation

The membrane bench-scale plant (Prozesstechnik GmbH, Basel, Switzerland), shown in Fig. 1, was provided with a non-stirred double-walled tank (maximum volume equal to 5 L) where the effluent (OMW/ST) was contained, and a diaphragm pump (Hydra-Cell model D-03) that served to drive the OMW/ST stream to a flat membrane module (M1), with dimensions of 3.9 cm width × 33.5 cm length.

The main operating variables were measured and displayed: the operating pressure was finely adjusted with a spring loaded pressure-regulating valve on the concentrate outlet (SS-R4512MM-SP, Swagelok) and monitored by a digital pressure gage (Endress + Hauser, model Ceraphant T PTC31), allowing independent control of the applied pressure ($P_{TM\ set\ point}$ ± 0.01 bar) and flowrate (0.1 L h^{-1} precision) that was

Table 1
Physicochemical characterization of OMW/ST.

Parameter	OMW/ST
pH	7.8–8.2
EC, mS cm^{-1}	3.2–3.6
TSS, mg L^{-1}	13.1–16.0
COD, mg O_2 L^{-1}	150.8–226.6
Total phenols, mg L^{-1}	0.4–1.0
Total iron, mg L^{-1}	0.03–0.4
Cl^-, mg L^{-1}	990.9–1045.1
Na^+, mg L^{-1}	534.1–728.7

OMW/ST: olive mill wastewater after secondary treatment.