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Experimental design optimization of reverse osmosis purification of pretreatedolive mill wastewater

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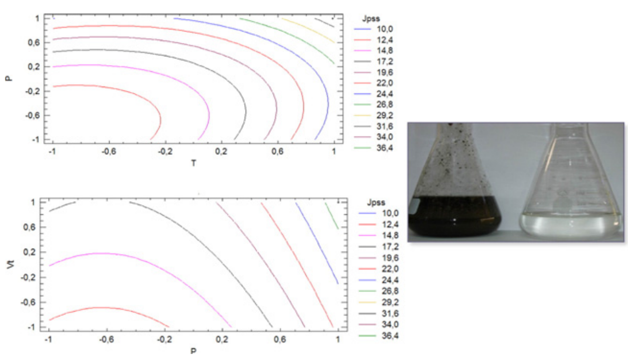
HIGHLIGHTS

- Reverse osmosis membrane (RO) process was optimized and modelled.
- Operating pressure (P_{TM}), crossflow (v_t) and temperature (T) influence.
- P_{TM} and T exhibit a deeper influence on the permeate flux than v_t.
- Optimum range 24-29.6 °C, 31.5-35 bar pressure and 4.1-5.1 m s^{-1} turbulent flow.
- Standards to reuse the purified effluent for irrigation or sewers discharge ensured.

GRAPHICAL ABSTRACT



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ABSTRACT

The management of the effluents generated by olive oil industries, commonly known as olive mills, represents an ever increasing problem still unresolved. The core of the present work was the modelling and optimization of a reverse osmosis (RO) membrane operation for the purification of a tertiary-treated olive mill wastewater stream (OMW2TT). Statistical multifactorial analysis showed all the studied variables including the operating pressure (P_{TM}), crossflow velocity (v_t) and operating temperature (T) remarkably influence the permeate flux yielded by the selected membrane (*p*-value practically equal to zero), confirming a statistically significant relationship among the variables considered at 95% confidence level. However, P_{TM} and T exhibit a deeper influence than v_t, according to the *p*-values withdrawn from the analysis, being the squared effects significant too, but more in case of the former ones. The obtained contour plots and response surface support the former results. In particular, the optimized parameters were ambient temperature range (24–29.6 °C), moderate operating pressure (31.5–35 bar) and turbulent crossflow (4.1–5.1 m s^{-1}). In the end, the quality standards to reuse the purified effluent for irrigation purposes and discharge to sewers were stably ensured.

1. Introduction

Within the last few decades the agro-industrial sector of olive oil has experienced a very significant expansion. As a consequence of this, the residues by-produced by these factories known in the jargon as 'olive mills' have also augmented considerably, in particular the generated wastewaters. The conversion of the production technology into continuous operation centrifugation processes in response to an ever more demanding market is the main responsible of this.

This fact has raised in the last years much concern in the producing countries, mainly in the Mediterranean Basin - Spain, Italy, Portugal,

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Greece and Northern African countries - where these agro-industries have represented historically an important engine of the economy. As an example of this, just in Spain there are over 1750 olive mills authorized and operating currently, representing around 45% of the worldwide production. Furthermore, this also concerns other regions where the production of olive oil has taken-off in the last years or is an emergent industry, which is the case of France, Serbia and Montenegro, Macedonia, Cyprus, Turkey, Israel, Jordan, Iran, and also the USA, Australia, and specially China, where it has strongly promoted recently and is expected to develop a considerable production potential in the near future (International Olive Oil Council, IOOC, 2013–2014). The management of the wastewaters from olive mills (OMW) is thus a task of global concern not anymore constrained to a specific region.

A series of hazards caused by these highly-polluted effluents in relation with the contamination of soil, deterrence of vegetation growth, pollution of water bodies and aquifers, inhibition of autopurification processes, phytotoxic impacts to the aquatic fauna and the ecological equilibria as well as strong odor nuisance have been reported so far (Danellakis et al., 2011; Karaouzas et al., 2011; Martínez Nieto et al., 2010; Niaounakis and Halvadakis, 2006; Ntougias et al., 2013).

It has been observed that conventional physico-chemical and biological processes as those based on activated sludge do not provide high effectiveness for the abatement of the organic load of OMW (Fountoulakis et al., 2002; Garrido Hoyos et al., 2002; Paraskeva and Diamadopoulos, 2006). The reason relies on the inherent physico-chemical characteristics of these effluents, mainly the acidic pH, high salinity, low alkalinity and nitrogen content and refractory organic contaminants, that make this wastewater potentially recalcitrant for whether aerobic or anaerobic treatments (Beltrán-Heredia and García, 2005; Sampaio et al., 2011).

A wide range of processes have been reported in the scientific literature up to now for the reclamation of OMW, but the vast majority have not been implemented at industrial scale due to cost-inefficiency or technical problems (Aktas et al., 2001; Al-Malah et al., 2000; Bouranis et al., 1995; Cegarra et al., 1996; Inan et al., 2004; Martínez Nieto et al., 2010, 2011a, 2011b; Papadimitriou et al., 1997; Paraskeva and Diamadopoulos, 2006; Tezcan et al., 2006; Stasinakis et al., 2008). Some researchers have highlighted co-digestion of OMW with agro-livestock as an apt solution (Gonçalves et al., 2012a, 2012b Kougias et al., 2014 Sampaio et al., 2011 Scoma et al., 2013). On another hand, advanced oxidation processes (AOPs), electrochemical and hybrid treatments have yielded the most promising results in terms of organic matter abatement and mineralization (Ammary, 2005; Beltrán-Heredia and García, 2005; Cañizares et al., 2007; Grafias et al., 2010; Inan et al., 2004; Lafi et al., 2009; Martínez Nieto et al., 2011b; Rizzo et al., 2008; Tezcan et al., 2006).

In this context, membranes have been a break-through in terms of advanced separation technologies: in first place, they are modular and compact, they are 'green' as separation can be achieved without addition of chemicals, and they are also significantly less intensive in terms of energy than conventional separation processes. Although membranes are a mature technology, many aspects are still in development and under investigation. In this regard, the main 'Achilles heel' is membrane fouling (Cheryan, 1998), investigated by a plethora of researchers in the last years in order to definitely convince investors to implement membranes as substitutes of a range of unit operations at industrial scale. In particular, in the field of wastewater treatments, this is especially problematic, given the low economic value of the product, that is, treated water (Iaquinta et al., 2009; Ochando-Pulido et al., 2013; Ochando-Pulido et al., 2014, 2015a, 2015b; Stoller and Bravi, 2010; Stoller, 2011).

Within this framework, the prediction of the performance of a selected membrane is mandatory for its operation when implemented in a treatment process at industrial scale (Stoller and Ochando Pulido, 2015). Concentration polarization and membrane fouling change dynamically during the operating time, making the output of the membrane unsteady. To solve this, a wrong decision taken by many engineers is to overdesign the membrane plant far too much, without taking into account the extent and nature of fouling in the particular process, strongly connected to the feedstock and membrane binomial (Stoller, 2011). This decision, though not unsuccessful to maintain the membrane performance, increases the costs and makes the process deficient.

The core of the present work was to model and optimize a reverse osmosis (RO) treatment process, at bench scale. For this objective, a factorial design was used for the optimization of the RO purification of a pretreated OMW stream (OMW2TT). The results were interpreted through the response surface methodology. A statistical multifactorial analysis was performed in order to quantify all the potential complex conjugated effects of the input parameters considered in the RO process. The process was subsequently modelled by means of a second-grade quadratic fitting model equation. Finally, the parametric quality standards that permit to reuse the purified stream for irrigation or discharge in-site reuse purposes were verified.

2. Experimental

2.1. Samples

Samples of OMW from different mills operating with the two-phase olive oil centrifugation extraction technology were collected during the winter campaign in the Andalusian provinces of Jaén and Córdoba (Spain). The samples were then taken to the laboratory and analyzed. OMW was thereafter conducted to a pretreatment process (OMW2TT) optimized and thoroughly explained in former research by the Authors (Martínez Nieto et al., 2011a, 2011b). The characteristics of the OMW2TT feedstream to the RO membrane unit are hereafter reported in Table 1.

2.2. Analyses

Analyses of the chemical oxygen demand (COD), total suspended solids (TSS), ashes, total phenolic compounds, total iron, electrical conductivity (EC) and pH were performed in both the influent (OMW2TT) and in the permeate of the RO unit, following standard methods (Greenberg et al., 2005).

A Helios Gamma UV–visible spectrophotometer (Thermo Fisher Scientific) was used for the analyses of the COD, total phenols and total iron. An ion chromatograph (Dionex DX-120) was used to measure the ionic concentrations. EC and pH were analyzed with a Crison GLP31 conductivity-meter and a Crison GLP21 pH-meter, provided with autocorrection of temperature (25 °C), previously calibrated with buffer standard solutions for EC (1413 μS/cm and 12.88 mS/cm) and pH (pH 4.01, 7.00 and 9.21) purchased as well from Crison.

Table 1
Physicochemical characteristics of OMW2TT.

Parameter	OMW2TT
pH	7.9 ± 0.2
EC, mS cm^{-1}	3.4 ± 0.2
TSS, mg L^{-1}	14.5 ± 1.5
COD, mg L^{-1}	188.7 ± 37.9
Total phenols, mg L^{-1}	0.7 ± 0.3
Total iron, mg L^{-1}	0.8 ± 0.3
Cl^-, mg L^{-1}	1018.0 ± 27.1
Na^+, mg L^{-1}	631.4 ± 97.3
HCO_3^-, mg L^{-1}	131.1 ± 1.8
Ca^{2+}, mg L^{-1}	93.9 ± 9.8
Mg^{2+}, mg L^{-1}	26.9 ± 5.6

OMW2TT: tertiary treated olive mill wastewater.