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Direct Contact and Vacuum Membrane Distillation application for the olive mill wastewater treatment

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ABSTRACT

In this work, Membrane Distillation (MD) in Direct Contact (DCMD) and Vacuum (VMD) configurations were applied for the Olive Mill Wastewater (OMWW) treatment, with the objective to obtain a purified stream to reuse, together with a concentrate rich in polyphenols. The experimental tests were carried out on capillary membranes using a membrane module realized in laboratory and equipped with three commercial polypropylene membranes (membrane pore size: 0.2 µm, thickness: 0.4 mm, inner diameter: 1.8 mm, membrane area: around 30 cm^2). The OMWW was frozen for its storage and, before experiments, simple filtration on single and multi-layer tissue were tested, to reduce membrane fouling during the process. The effect on the permeate flux and purity of the feed temperature (30–40–50 °C) for the DCMD and of the feed concentration for the VMD, was investigated.

1. Introduction

The olive oil extraction by means of continuous and automatic centrifugal systems, although usually leads to higher efficiencies and better oil quality than the traditional hydraulic presses, significantly increases the quantity of wastewater to be managed/disposed. The produced OMWW are often spilled directly into the ground [1] causing environmental problems as soil contamination and water pollution, because of the high amount of phenolic compounds contained in the discharged residues. Nevertheless, the recovery of polyphenols is of high interest, due to their important benefits for human health. In this scenario, the development of technologies for the purification of OMWW and the recovery of polyphenols has considerably increased in last years. Membrane separation techniques are well known for their ability to separate, concentrate and purify chemical species from liquid solutions, showing several advantages with respect to traditional operations, like high selectivity, low energy consumption, possibility of developing integrated systems. Moreover, membrane technology is considered a powerful tool for the sustainable industrial development, being able to well respond to the goal of the “process intensification strategy” in terms of reduction of the plant size, increase of the plant efficiency, reduction of energy consumption and environmental impact. To date, several membrane operations were studied for the treatment of OMWW such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO), also in integrated configurations [2–7]. These operations represents the “traditional” class of membrane systems, and are well consolidated in different fields, like the seawater desalination for the production of drinking water, the treatment of effluents of dairy industry, etc.

More recently, Membrane Distillation (MD) was also studied in various sectors of industrial interest: treatment of waste water from textile processes, purification of waters containing arsenic, treatment of salty solutions, concentration of acid solutions; treatment of radioactive waste, drying of solid microparticles from aqueous suspensions, etc. [8–17]. The use of low temperatures makes the process also compatible with the needs of the food industry: it is possible, for example, to concentrate fruit juices improving flavor and color of the final product [18,19]. Furthermore, working MD at atmospheric pressure, the feed is processed under milder condition than the pressure-driven membrane operations and fouling issues, as well as the problem of membrane compaction, are reduced.

In the literature, few works on the use of MD for OMWW treatment, in direct contact (DCMD) configuration and using flat-sheet membranes, have been reported. The tested membranes were in PTFE and PVDF. In Table 1, the obtained flux and rejection values are summarized. A preliminary study of vacuum membrane distillation (VMD) in an integrated MF-NF-VMD membrane system was also presented by Garcia-Castello et al. [20] who just measured the

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Table 1
Literature data on DCMD applications for OMWW treatment.

Flat membrane characteristics	Operating conditions	Main results	Ref.
PTFE (TF200, Gelman) dp[a]: 198.96 nm thickness: 55 ± 6 μm LEPw (10^5 Pa): 2.76 ± 0.09 Porosity: 69 ± 5% PVDF (GVHP, Millipore) dp[a]: 283.15 nm thickness: 118 ± 4 μm LEPw (10^5 Pa): 2.04 ± 0.03 Porosity: 70 ± 3%	Feed: OMW (Marrakech Morocco) 2006/2007 season No pretreatment T_{feed} = 40°C T_p = 20 °C Stirring rate = 500 rpm	TF200 J(t = 0) = 7.68 ± 0.22 L m^{-2} h^{-1} GVHP J(t = 0) = 4.954 ± 0.14 L m^{-2} h^{-1} TF200 α[b](t = 9 h) = 99% GVHP α[b] (t = 9 h) = 89%	[21]
PTFE (TF200, Gelman Science) supported by a polypropylene net dp[a]: 0.2 μm thickness: 178 μm (support included) porosity: 80% LEPw (10^5 Pa): 2.76 Membrane area: 2.75×10^{-3} m^2	Feed$_1$: crude OMW Feed$_2$: OMW with coagulation/flocculation as pre-treatment Feed$_3$: OMW with microfiltration (MF) as pre-treatment T_{feed} = 30–60 °C T_p = 20 °C Stirring rate = 500 rpm Initial volume of feed and permeate chambers: 0.25 L	*Considering MF as optimal pretreatment* J (t = 0; T = 40 °C) = 7.7 L m^{-2} h^{-1} α[b] (t = 8 h; T = 40 °C) = 99% C_p (t = 8 h; T = 40 °C) = 50 ppm *Long term experiment* (40 °C) J(t = 20 h; T = 40 °C) = 2.5 L m^{-2} h^{-1} J(t = 30–76 h; T = 40 °C) = 1.44 L m^{-2} h^{-1} C_p (76 h) = 0.2 g/L *Fouling study* $Jw_{initial}$ = 9.7 L m^{-2} h^{-1} $Jw_{after\ tests\ on\ OMWW}$ = 6.3 L m^{-2} h^{-1} (Jw reduction = 35%) J(t = 0;T = 50 °C) around 10 L m^{-2} h^{-1} α[b] (t = 8 h; T = 50 °C) = 100%	[22]
PTFE polymer supported by a polypropylene net (Gelman) TF200 dp[a]: 0.2 μm thickness: 178 μm effective porosity: 80% LEPw = 282 kPa TF450 dp[a]: 0.45 μm thickness: 178 μm effective porosity: 80% LEPw = 138 kPa TF1000 dp[a]: 1.0 μm; thickness: 178 μm effective porosity: 80% LEPw = 48 kPa Membranes area: 2.75×10^{-3} m^2	Feed: OMW from a three-phase olive mill unit located in the region of Marrakech (Morocco) T_{feed} = 40–50 °C T_p = 20 °C stirring rate = 500 rpm	TF200 J(t = 0;T = 40 °C) = 7.68 ± 0.22 L m^{-2} h^{-1} α[b] (t = 8 h; T = 40 °C) = 99.5% TF450 J(t = 0;T = 40 °C) = 8.26 ± 0.24 L m^{-2} h^{-1} α[b] (t = 8 h; T = 40 °C) = 99.1% TF1000 J(t = 0;T = 40 °C) = 8.84 ± 0.25 L m^{-2} h^{-1} α[b] (t = 8 h; T = 40 °C) = 98.7%	[23]

[a] *dp* = mean pore size.

[b] α = polyphenols separation coefficient ($\alpha = 1 - C_P(t)/C_0$), with C_p and C_0 the polyphenol content in the permeate and the feed, respectively.

trans-membrane flux achievable with this configuration operating on the NF permeate at low feed temperatures: at 30 °C, P_p = 30 mbar; Q_{feed} = 180 L/h and with a PVDF flat membrane (Membrane area: 55 cm^2; dp: 0.2 μm; thickness: 200 μm), the initial flux registered was of 11 L m^{-2} h^{-1}.

In the present work, the potentialities of the MD process, in both DCMD and VMD configurations, were investigated for the OMWW treatment. For the first time, polypropylene (PP) capillary membranes were chosen. PP is a material employed in many industrial fields, and the study of its behavior when in contact with the OMWW is essential to establish a possible use also in this specific sector. Commercial capillary PP membranes were, then, tested in a lab-assembled module. This membrane configuration was chosen because, as well known, the use of capillary membranes allow to work with higher area/volume ratios than the flat configuration and, then, with smaller units, as requested by the process intensification strategy. OMWW may alter its composition with time and, therefore, its storage represents and important step in the whole treatment procedure. It has to be noticed that there is not an established storage method to follow in order to avoid variations in the feed composition. For example, El-Abbassi et al. [21,22] stored OMWW samples at room temperature, whereas Russo [6] acidified from pH 5.5 to 3.5 to prevent phenols oxidation. In both cases, compositions changes may occur. Moreover, the acidification step implies the use of chemicals inside the plant. In this work, OMWW samples were frozen as collected. The OMWW freezing can also affect the feed composition and, therefore, should be considered as a sort of pre-treatment step. MD tests were, then, carried out using the thawed feed, after its filtration through a single/multi layer tissue. To evaluate the potential of PP membranes for the OMWW treatment, the performance of DCMD and VMD was investigated in terms of permeate flux and rejection values, in order to check the possibility to obtain a purified permeate, together with a high concentrated retentate rich in polyphenols.

2. Materials and methods

2.1. Feed samples

The OMWW feed was kindly provided by “Frantoio VELTRI” (Longobardi, Cosenza, Italy). The samples were frozen at their arrival for storage and thawed after for testing. In particular, pretreated waste streams were employed for carrying out the experiments in order to reduce fouling phenomena during the process, and, consequently, to increase the values of permeate fluxes as well as to minimize the time of membrane cleaning after each test. A simple filtration at ambient temperature by a single (F1) and eight layers-tissue (F8) was used to remove suspended particles from the feed. The pictures (realized with digital microscope-Axiovert 25) of single and eight layers-tissue are shown in Figs. 1 and 2, respectively. For the single layer tissue it was possible to calculate the mesh size (average area of 3565 μm^2), while the multi-layers tissue did not present any clear mesh, being obtained by overlapping eight single layers.

2.2. Membrane properties and membrane module preparation

The experimental tests were carried out using commercial capillary membranes made of polypropylene (average pore size of 0.2 μm; thickness of 0.4 mm; porosity around 70%; inner diameter of 1.8 mm) purchased by Membrana (Germany).

The membrane module was prepared by inserting three capillaries in a glass device, sealing both extremities with epoxy glue. The membrane area was around 30 cm^2.