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Ceramic macromembrane for tangento-axial micro- and ultrafiltration water systems

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ABSTRACT

An innovative proposal of tangento-axial filtration based on the concept of macromembrane (large size) is presented in the context of micro- and ultrafiltration ceramic membranes. The macromembrane is made of Al_2O_3–TiO_2 by slip casting. It includes an internal system for rotating the fluid trough a propeller which enhances the tangento-axial filtration. This filtration yields better than the cross-flow filtration in conventional extruded ceramic tubes of small diameter. The reasons are, first, its larger sizes relative to the extruded tubular filters, and second, that it can work at higher pressures, resulting in better operational performance. Furthermore, costs of industrial-scale production could be lower. In this study, the conceptual basis, pilot plant, first experimental results, and proposals for improvement of the system to continue the project are presented.

Keywords: Ceramic macromembrane; Tangento-axial filtration; Microfiltration; Ultrafiltration; Water treatment

1. Introduction

During the last 20 years, there have been significant advances on systems for filtration of fluids including water for both wastewater reuse and human consumption [1]. Filtration systems are based on organic (usually polyamides) or inorganic (ceramic) filters. It should be noted that ceramic membranes allow conditions of use at high temperatures (above 200˚C) as well as cleaning processes in extreme pH conditions (<3 and/or >12) to which organic filters cannot stand. Hence, they have higher levels of use and durability than those of organic filters [2,3].

There are two types of ceramic membranes in the market: the tubular membranes, obtained by extrusion of ceramic slurries (cross-flow technology), and the flat membranes used in total filtration and obtained by pressing a ceramic green paste before sintering.

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Alternatively, in this work, a ceramic membrane was produced by slip casting from ceramic slurry. This technique allows us to obtain membranes of any shape and large size, so that henceforth they will be called "macromembranes." Thanks to its large size, these macromembranes allow to host on its interior devices, such as propellers, and thus achieve a tangento-axial flow that helps to reduce the formation of fouling on their inner wall.

The slip casting technique consists in preparing a water-based slurry of high-density water-insoluble particles of Al_2O_3–TiO_2 with the support of auxiliary products such as dispersants, defloculants, additives, and release agents [4,5]. In this method, the process of obtaining the macromembrane takes place in four phases: (i) preparation of the slurry to control viscosity and thixotropy, (ii) putting it into a polymer mold or molding plaster, (iii) demolding and drying, and (iv) sintering by an adequate thermal curve (maximum temperature equal to 1,300˚C). Details about the description of the ceramic process can be found in [6].

Following this production process, large ceramic macromembranes with different shapes were obtained. Those with frusto-conical shape and the following dimensions: height 500 mm; higher diameter 400 mm; smaller diameter 40 mm; and thickness 8 mm were chosen (Fig. 1).

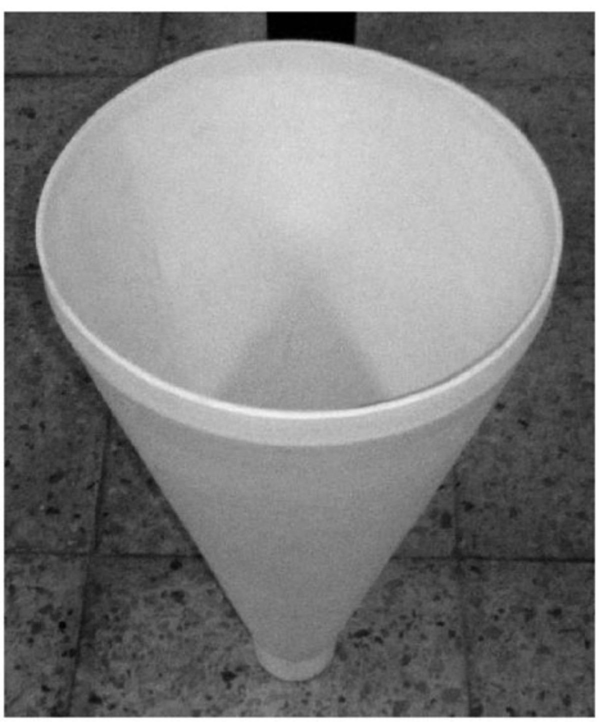

Fig. 1. Ceramic macromembrane made of Al_2O_3–TiO_2: maximum inner diameter, 400 mm; minimum inner diameter, 40 mm; height, 500 mm; thickness, 8 mm.

2. Why ceramic macromembranes?

Separation processes based on membranes are having a progressive level of development in recent years due to high oil prices, such as membrane distillation. It is a process based on the vapor transport across a hydrophobic microporous membrane driven by the vapor pressure gradient across the membrane [7]. However, one of the main problems is the degradation suffered by these membranes over time. In contrast, ceramic membranes do not have this problem [8,9].

In the case of large-size membranes, it is possible to increase the yield in filtration due to the fact that some new processes can be incorporated inside them: electrocoagulation, magnetic fields, catalysis, ultrasound, gaseous diffusion, and mixing.

In contrast to extruded tubular membranes, these macromembranes allow us to create a rotational flow by incorporating stirrers and/or baffles, therein that create a dynamic pressure on their inner walls (Fig. 2). The conical shape allows a dragging effect that decreases fouling on the walls by both generating a tangento-axial pressure on the macromembrane and by driving the self-cleaning of it.

The permeate flow is subjected to a transmembrane pressure described by the Darcy's law [10]:

Fig. 2. Experimental stirrer used in pilot tests.

$$q = -K(\Delta p)^n; \quad n \neq 1 \tag{1}$$

$$K = k\,\mu/\gamma \tag{2}$$

where K = Darcy's permeability (it depends on the porosity and fluid), Δp = transmembrane pressure, k = intrinsec permeability (it depends on the porosity), γ = specific weight of the fluid, and μ = dynamic viscosity of the fluid (it depends on the fluid temperature).

On the other hand, centripetal pressure in a fluid inside a circular cylinder of radius r is

$$\Delta p = {}^1\!/\!_2 \rho \omega^2 r^2 \tag{3}$$

where ω is the rotational speed of the fluid and ρ is the density. Assuming a purely Newtonian fluid in turbulent regime, the exponent $n = 2$ in Eq. (1) can be considered valid, in contrast to the case of laminar flow ($n = 1$) which would correspond to conventional cross-flow filtration. Then, the permeate flow would be approximately

$$q \approx \omega^4 r^4 \tag{4}$$

That is why the purpose of this research work aims to achieve operational yields in ceramic filtration processes far superior to those achieved by conventional cross-flow filtration [6].

3. Experimental

The approximate formulation used to prepare the slurry of the macromembranes is as follows: (percentages by weight): corindon, 26%; alumina, 31%; rutile, 4%; glycerin, 6%; dextrin, 5%; ball clay, 5%; water, 22%; other compounds, 1%. This is a slurry made from non-plastic materials. Hence, it is necessary to grant the appropriate plasticity, viscosity, and thixotropy to obtain the workpiece, through a mold prepared with special plaster, to which a thin layer of mold release product is adhered.

Obtaining the membrane requires a preparation of products in ball mill and careful drying environmental conditions. Once produced the ceramic macromembrane, a pilot plant has been built consisting of the following elements (Fig. 3):

(1) ceramic macromembrane based on Al_2O_3–TiO_2 spinels;
(2) outer conical casing made of stainless steel for holstering the macromembrane; it includes a polycarbonate window for inspection purposes;
(3) propeller for stirring the fluid, including a stirrer and a electric motor;
(4) two thermally insulated tanks for storage of fluid and stirring at different operating temperatures;
(5) pump that allows fluid pressure up to 7 bar;
(6) controller for controlling the pressure of the pump, the rotational speed of the propeller, and the fluid temperature;
(7) compressor for cleaning the macromembrane through countercurrent air pulses.

Fig. 3. Pilot plant for testing macromembrane performance.

Assessment of porosity or "cut off" pore size of the experimental macromembrane was obtained by electron microscopy and mercury porosimeter, giving values of 1–2 μm in the membrane support before applying the active filter layer of micro and/or ultrafiltration (Fig. 4).

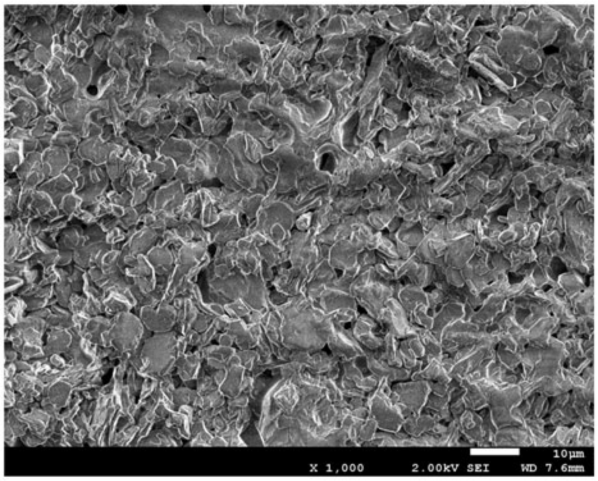

Fig. 4. Picture by SEM showing the microstructure of the Al_2O_3–TiO_2 membrane body. Pore sizes are 1–2 μm, approximately.

4. Results and discussion

The tangento-axial filtering system has been tested by using tap water as test fluid. The system has been subjected to different conditions of inlet fluid temperature, pressure, in and out flowrate, and rotational speed of the stirrer.

Permeability and hydraulic conductivity ranged between 145 and 653 L/(m^2 h) and 148 and 545 L/(m^2 h bar), respectively, depending on operating conditions (Table 1). These results are so far below than expected. The reasons that explain this poor performance may include the following, among others:

(1) The first replica of the frusto-conical macromembrane broke at 2.5 bars of pressure when tangential forces were applied (Fig. 5). For safety reasons and after several experiments that ended with cracks in the walls of the macromembrane, the subsequent tests were conducted at low pressure (0.5—1.8 bar). Although, an increase in pressure would increase the permeability;

(2) The effect of the stirrer was tested by varying rotation speed (from 0 to 125 rpm) and direction of rotation (counterclockwise and clockwise). For the fluid used (water) effects on permeability were irrelevant (Fig. 6). A new design of the stirrer and baffles could result in a substantial improvement of the system. However, it has been found effective in reducing the effects of fouling inside the macromembrane. In the case of filtering high viscosity fluids the system should provide results where the utility of its function of tangento-axial filtration would become more evident.

Table 1
Some data and results of the tests carried out on the pilot plant

Macromembrane inner diameters	400/40 mm
Pore size	1—2 μm
Apparent porosity	34%
Density of the ceramic material	2.27 g/m^3
Macromembrane length	500 mm
Inner surface	0.37 m^2
Macromembrane volume	23 L
Macromembrane thickness	8 mm
Working pressure	0.5—2.5 bar
Inlet temperature	17—44℃
Input flowrate	890—1,050 L/h
Stirrer rotation speed	0—125 rpm
Permeate flowrate	217—578 L/h
Permeability	145—653 L/(m^2 h)
Hydraulic conductivity	148—545 L/(m^2 h bar)

Fig. 5. Picture of the macromembrane broken at 2.5 bars. The stirrer can be seen in the interior of the macromembrane.

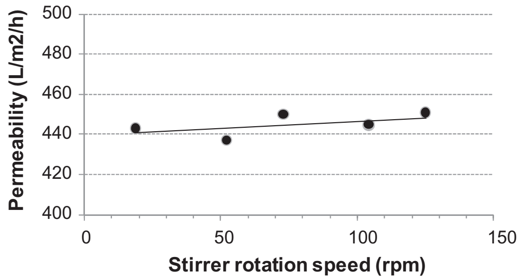

Fig. 6. Effect of stirring in the performance of the macromembrane. Operating conditions: inlet temperature = 18˚C; inlet flowrate = 890 L/h; pressure = 1.5 bar; duration of each measure = 10 min.

The frusto-conical shape of the membrane had breakage problems at pressures above 2.5 bar. This was mainly due to the large tangential pressures that the macromembrane bears on its top (larger diameter). For a conical vessel, this circumferential stress, i.e., the normal stress in the tangential direction of the macromembrane, is given by the following equation [11]:

$$\sigma_\theta = \frac{p\,r}{e\,\cos\,\alpha} \tag{5}$$

where σ_θ = circumferential stress or hoop stress (bar), p = pressure (bar), r = radius (m), e = thickness (m), and α = semiangle in the apex of the cone.

By applying Eq. (5) to the macromembrane, with $\alpha = 19.8^\circ$, the calculated stress is 66 bar, approximately. A bend-breaking test of the ceramic material gave a stress of 820 bar with a standard deviation of 70 bar. Therefore, in future developments it seems to be necessary to strengthen the top with a greater thickness or by ribs. In the current state of the research, this is a handicap for the experimental verification of the model that predicts a large increase in the permeability at elevated pressures.

The pilot tests lasted for more than 24 continuous hours without noticing an important decrease of permeability. As example, Fig. 7 shows the variation of the permeability over time, maintaining constant operating conditions. The reduction in permeability was 20% approximately in 24 h. In comparison, this percentage is much lower than those observed in the case of tubular ceramic membranes, which ranged between 28 and 74% according to a study in which several types of membranes were compared [12]. Furthermore, after 24 h, the permeability of the macromembrane remained in 378 L/(m^2 h), while that in the case of the tubular membranes was reduced to 225–285 L/(m^2 h) [12]. So that, even at this initial stage of the project, the process of self-cleaning through tangento-axial drag in the macromembrane seems to meet design expectations.

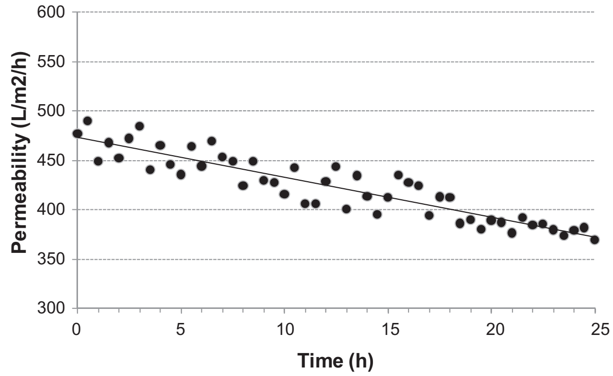

Fig. 7. Performance of the macromembrane in the pilot plant. Permeability vs. time at constant operating conditions: inlet temperature = 20˚C; inlet flowrate = 890 L/h; pressure = 1.5 bar; no stirring.

As far as economic aspects are concerned, costs of the macromembranes prepared by slip casting would have advantages over the tubular membranes for cross-flow filtration, for the same filtration surface, regardless of the fact that they could integrate into a single-system additional processes than conventional technology cannot incorporate due to spatial limitations already known. Capital cost of a frusto-conical membrane of 0.6 m^2 surface is approximately 160 €/unit, i.e., 267 €/m^2. The cost of a commercial tubular ceramic membrane is around 300–600 €/m^2 [13]. On the other hand, the energy costs in the macromembrane system could be higher because it is necessary to consider the energy to activate the internal devices that the macromembrane can lodge. However, the energy costs associated with back-flushing could be lower due to the self-cleaning effect of tangento-axial flow.

5. Conclusions

A new filtration process using ceramic macromembrane has been introduced, which applies the concept of tangento-axial filtering vs. conventional cross-flow filtration. It should be mentioned that the system can work at high temperatures in the presence of aggressive chemicals (strongly acidic or basic pH) and a variety of operating conditions and *in situ* mixing.

Possible applications in the treatment of fluids include the following main sectors: (i) biotechnology and pharmaceutical industry; (ii) food industries (dairy, wine, juices, etc.); (iii) comprehensive water purification treatments; (iv) recovery of industrial effluents (oils, lubricants, etc.) and; (v) integrated chemical reactors, etc.

In the present state of research, several challenges must be overcome before being able to do the technology transfer to the industrial production: (a) improving the ability to withstand the pressure of the fluid, (b) redesigning and testing new stirrers, and (c) experimenting with viscous or high density fluids.

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