APPLICATION OF SILICON CARBIDE HOLLOW FIBER MEMBRANES IN THE PURIFICATION OF BIOACTIVE COMPOUNDS FROM CAGAITA JUICE

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ABSTRACT

The application of membrane separation processes in the separation and purification of bioactive compounds present in natural products has been widely used. Ceramic membranes have greater thermal, chemical, and mechanical resistance, when compared to polymeric membranes. Furthermore, hollow fibers provide a greater filtration area per volume occupied, when compared to other geometries. Ceramic membranes of the hollow fiber type can be produced by the inversion method phases followed by sintering. This method generating different structures and distribution of pore sizes. Silicon carbide is a ceramic material widely used in the literature for manufacturing ceramic membranes. Cagaita is a cerrado fruit that has high nutritional, functional and economic potential, but is still little explored in the literature. Phenolic compounds are widely known for their antioxidant and anti-inflammatory properties associated with health benefits. In this study, ceramic membranes of the hollow silicon carbide fiber type were produced using the phase inversion method followed by sintering, and applied to the separation of bioactive compounds present in cagaita juice.

Keywords: Hollow fiber. Membranes separation processes. Bioactive compounds. Cagaita.

1 INTRODUCTION

The application of ceramic membranes in separation processes has stood out in several areas, such as water treatment[1], gas separation[2], heavy metal removal[3] and for the concentration of natural extracts in the food industries[4]. The advantages of hollow fiber over other geometries are the high ratio surface area/volume. The phase inversion method followed sintering can be applied to manufacture of ceramic hollow fiber membranes with asymmetric structures.

The silicon carbide is widely used in literature for fabrication of ceramic membranes[5,6]. The sintering of silicon carbine requires sintering temperature superiors of 2000 °C due to strong covalent bonds at material. In addition, the heat treatment should be carried out in an inert atmosphere for prevent oxidation. Due to these limitations, some studies propose the use of sintering aids, such as silica[1], zeolite[7] and others.

Cagaita (*Eugenia dysenterica*) is the small fruit, with greenish yellow peel, juicy and acidic pulp typica on the region from cerrado and little explored in the literature. The literature shows that the cagaita presents high economic, nutricional and functional potencial[8]. Phenolic compounds are widely known for their antioxidant and anti-inflammatory properties associated with health benefits, such as cancer prevention and obesity reduction. Antioxidants, antiobesity and antidiabetic properties of polyphenolic compounds have been investigated by pharmaceutical and food industries[9].

In this study, ceramic hollow fiber membranes were produced by the phase inversion method followed by a sintering step. The ceramic material used was silicon carbide and silicon were the sintering aids. The membranes were analysed for mechanical resistance, hydrophilicity, scanning electron microscopy and water permeability. Filtration was carried out in cross-flow and pH, turbidity and compounds phenolic in the feed and in the permeate were quantified.

2 MATERIAL & METHODS

2.1. Material

Silicon carbide (SiC – $D_{0,1}$ = 3,50 µm; $D_{0,5}$ = 12,15 µm; $D_{0,9}$ = 24,97 µm) and silicon (Si – $D_{0,1}$ = 2,18 µm; $D_{0,5}$ = 6,19 µm; $D_{0,9}$ = 15,93 µm), supplied by RIMA Industrial S/A (Brazil), were the ceramic materials used in composition of fibers. Polyethersulfone (PES, Veradel 3600 P, Solvay) and dimethyl sulfoxide (DMSO, Vetec, Brazil) were used as binder and solvent, respectively, for preparing the ceramic suspension. Total phenolic compounds were determined using the following reagents: Folin & ciocalteu´s phenol reagent, Sigma; Tannic acid (purest), Vetec; anhydrous sodium carbonate P.A. (Na2CO3), Vetec.

2.2. Fabrication of hollow fiber membranes

The hollow fibers were manufactured using the phase inversion method followed by sintering as recommended by literature[10]. The ceramic suspension was composed by 30 wt% of ceramic material, 60 wt% of solvent (DMSO), 0.65 wt% of additive (Arlacel) and 9.35 wt% of polymer (PESf). The ceramic material was composed of 70 wt% of silicon carbide and 30 wt% of silicon. The final ceramic suspension was degassed in a vacuum pump for approximately 2 h for air bubbles removal. In the extrusion stage the ceramic suspension was extruded through a tube-in-orifice spinneret (outer diameter of 3 mm, inner diameter of 1.2 mm) into a tap water bath, using two individual pumps (Harvard Apparatus, model XHF). A mixture formed by ethanol (30 wt%, Sigma Aldrich, 99,8%) and DMSO (70%) were used as internal coagulants. The air-gap distance was fixed at 0 mm. Flows of the internal coagulant and ceramic suspension were controlled at 25 and 15 mL min^{-1}, respectively. Sintering was carried out in a tubular furnace (Carbolite model TZF 15) and the ramp was based in literature[10].

2.3. Characterizations

The morphological structure of the fibers was evaluated by scanning electron microscopy (SEM) using the Carl Zeiss microscope, model EVO MA 10. Image J was used to evaluate the dimensions of the fibers in the images obtained with the SEM. The mechanical strength of the fibers was measured by the three-point bending test using the Instron Model 9600 equipment coupled with a 5 kN cell and using hollow fibers with a length of 30 mm[10].

Filtration and water permeability tests were carried out at room temperature (28 ± 3°C) an in *cross-flow* operating mode. The cagaita juice was prepared in a 1:8 weight cagaita/water. The cagaita and water were crushed, and the juice was pre-treated with sieving (Bertel Industrial Metallurgic, opening of 53 mm/μm and mesh of 270). The permeate in the sieve was used to feed the filtration carried out with the silicon carbide hollow fiber. Filtration occurred at a pressure of 1 bar, using a feed volume of 350 mL and a filtration area for $4.12.10^{-4}$ m^2. Turbidity analyses were performed on a TB-200 digital benchtop turbidimeter. Total phenolic content was quantified by the Folin-Ciocalteu method using a standard tannic acid curve (R^2=0,995) and the absorbance was measured at 760 nm (UV – Shimadzu). The results were expressed in mg of tannic acid equivalent per g of juice. The retention of phenolic compounds was calculated in relation to the contend of phenolic compounds in the membranes feed.

3 RESULTS & DISCUSSION

The SEM images of the cross-section and external surface of the membrane produced from silicon carbide, sintered at 1350°C, are shown in Figure 1. In the cross-section of the fiber (Fig. 1 (a)) it was possible to observe asymmetric pore size distribution with micro-channels formed from the outer surface and spongy structure was detected throughout the central region of the fiber and in the vicinity of the inner surface. During the phase inversion process, the solvent leaves the fiber and non-solvent (water) enters, thus, this displacement of solvent (DMSO) and non-solvent (water) promotes the formation of filaments. As the fluid used as an internal coagulant was a mixture of solvents, this exchange occurred only from the external surface, which justifies the non-formation of filaments from the internal surface.

The microporous region of the fiber formed from the outer surface of the fiber is equivalent to 41.36% of the total cross-sectional region of the fiber. Reports in the literature using these same extrusion parameters but with different ceramic materials show the achievement of different porous structures[11]. The difference in the distribution of filaments in the fiber structure may be related to the different viscosities observed for each ceramic suspension resulting from different ceramic materials.

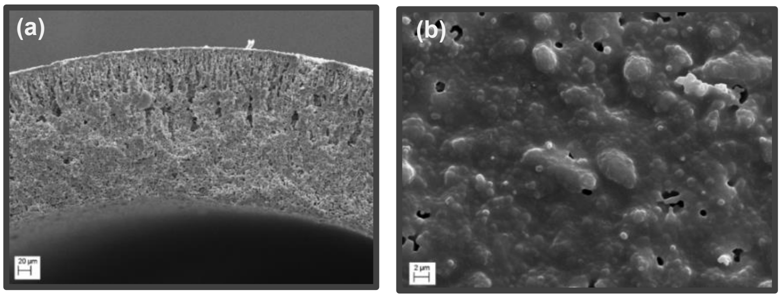

Figure 1 SEM images of the (a) cross-section e (b) outer surface of the hollow fibers sintered at 1350 °C.

The mechanical strength and permeability water of the silicon carbide hollow fiber were 57,04 ± 11,93 MPa and 9.52 L h^{-1} m^{-2} kPa^{-1}, respectively. These characterizations are in accordance with membranes discussed in the literature to be applied in processes involving microfiltration[5,7]. Furthermore, the presence of the sintering aids allowed the fiber to be sintered at temperatures lower than 2000 °C.

This hollow fiber was used in the filtration of cagaita juice and the turbidity, pH and retention of total phenolic compounds were evaluated. Figure 2 shows the experimental flow obtained during filtration. Furthermore, the flows obtained with the adjustment of the model proposed by Field[12] to evaluate the occurrence of fouling during filtration. The initial experimental flow was approximately 225 L h^{-1} m^{-2}. This value is approximately 4 times lower than the water flow through the membrane. The most pronounced decline in flow occurred in the first 6 minutes of filtration followed by stabilization at approximately 47 L h^{-1} m^{-2} after 50 minutes of filtration. Analyzing the adjustment made with the experimental data as proposed by the Field model, the formation of cake and partial blockage of the pores can be seen, adjustments that best represented the experimental data.

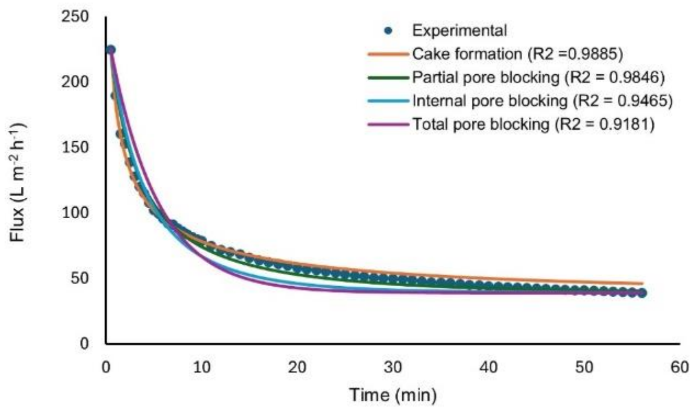

Figure 2 Experimental and calculated flux data for filtration of cagaita juice.

The permeate and feed samples were analyzed for turbidity, pH and total phenolic compound content. The pH of the cagaita juice was approximately 3.35 ± 0.1 both feed and permeated. The filtration decreased the juice turbidity by 97.75%, and the permeate samples was visually clearer than the feed sample. Regarding the concentration of total phenolic compounds, there was a reduction in the content of these compounds by 57.2%. The literature has already reported the application of hollow alumina fibers applied in the filtration of green tea extract to obtain phenolic compounds, with a reduction in turbidity of 90% and a rejection rate of phenolic compounds content of 10%[13]. The literature has also evaluated the application of flat polymeric membranes in the retention of total pequi polyphenol compounds, with a polyphenol retention rate in the range of 45%, using sequential filtrations[9].

4 CONCLUSION

The hollow fiber type ceramic membranes produced with silicon carbide and silicon as a sintering additive, presented mechanical resistance and water permeability with values suitable for application in microfiltration processes. Furthermore, they were obtained using sintering temperatures lower than those recommended in the literature. The application of these fibers in the filtration of cagaita juice provided a reduction in juice turbidity of 97.75% and a retention of total polyphenol compounds of 57.2%. These membranes proved to be efficient for application in the clarification of cagaita juice and retention of total phenolic compounds.

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ACKNOWLEDGEMENTS

The authors gratefully acknowledge Brazilian funding agencies (CNPq, CAPES, FAPEMIG, FAPDF) for financial support.