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Stability of oil-in-water emulsions produced by membrane emulsification with microporous ceramic membranes

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ABSTRACT

Membrane emulsification has been drawing attention due to its many application possibilities. This study aimed to assess the preparation and stability of oil in water (O/W) emulsions by membrane emulsification technique. Microporous ceramic membranes with mean pore size 0.2 μm and 0.8 μm were used for producing sunflower oil in water emulsions, displaying mean droplet size ranging from 2.9 to 11.6 μm and from 4.8 to 16.2 μm, respectively. The effect of the oil concentration (10%–20%), surfactant concentration (1%–4%), tangential velocity (0.12 m s^{-1} to 0.24 m s^{-1}), and feed pressure (100 kPa–300 kPa) on the process performance was investigated using an experimental design. All the parameters significantly influenced the mean droplet size, the droplet size distribution, and the emulsion stability. Depending on the emulsification conditions, monodisperse and polydisperse emulsions were obtained. The use of a membrane with 0.2 μm mean pore size led to better results than those obtained with a 0.8 μm membrane, since they yielded emulsions displaying smaller droplet size and narrower size distribution. The emulsions prepared using this membrane were stable up to 100 days.

1. Introduction

Emulsions play an important role in many industrial applications and products such as cosmetics, pharmaceuticals, and foods (Piacentini et al., 2014). Yet, emulsification processes have not changed much until the last decades, when membrane emulsification (ME) technology started to be developed. This method represents an alternative way to produce emulsions and particles by permeating the disperse phase, e.g. a vegetable oil, through a porous membrane to form a drop-by-drop emulsion. These droplets detach from the membrane surface due to the shear stress of the moving continuous aqueous phase. To maintain the stability of the emulsion and avoid the coalescence of the droplets, one or more surface active agents are generally used (Imbrogno et al., 2014; Matos et al., 2013; Piacentini et al., 2014).

Amongst the benefits of ME technology are the control of the droplet mean size and size distribution by selecting the membrane with specific pore size, reduced energy requirements, highly flexible manufacturing plant, use of mild operation conditions, and multiple possibilities for manufacturing simple and multiple emulsions, microspheres, and liposomes (Piacentini et al., 2014). Furthermore, another advantage relies on the suitability of membrane systems for large-scale production, mainly because they are based on the multiplication of membrane module processing units (Laouini et al., 2013). ME techniques can be divided into direct ME, when the disperse phase is injected through the membrane into the continuous phase, and premix ME, when a coarse emulsion is pressed through the membrane to reduce the droplet size.

Membranes used in the membrane emulsification process are specifically designed for this purpose, such as the Shirasu Porous Glass (SPG) membranes, due to their narrow pore size range. Besides, ceramic membranes have also been used for ME in cross-flow systems (Matos et al., 2013). An increasing interest in the different applications of ceramic membranes has been observed (Emani et al., 2014; Khemakhem et al., 2013; Matos et al., 2013). Ceramic membranes have some advantages in water-oil separation, such as mechanical and thermal resistance, chemical inertness and non-

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swelling, i.e., less interaction between the solvent-solute-membrane. In addition, the oil that adheres to the ceramic membrane can be easily removed by heat treatment for recovering the membrane permeate flux. Studies performed by Matos et al., (2013), using commercial available microfiltration flat and tubular ceramic membranes, resulted in the formation of O/W emulsions displaying narrow size distribution, as assigned by the very low span values, and only a slight influence of the shear stress on mean droplet diameter and size distribution was verified. In addition, the differences in the droplet size verified using flat and tubular membranes were associated with pore activation by the dispersed phase pressure. Emani et al. (2014) evaluated the effect of some operating parameters (cross flow velocity/Reynolds number and transmembrane pressure differentials) in the treatment of oil-in-water emulsions using tangential microfiltration with low cost ceramic membranes and high feed concentrations (400–500 mg/L). Low fouling index during microfiltration process was observed, suggesting greater membrane lifespan and consequently lower industrial costs. Chang et al. (2014) used nano-TiO_2 modified commercial ceramic microfiltration membranes in the preparation of stable oil-in-water emulsions. The modified membrane showed better performance than that of the unmodified membrane due to the hydrophilic coating, which, in turn, avoided the adhesion or penetration of the oil droplet by deformation, contributing to reduce the membrane fouling. The optimization of the operational parameters allowed to the improvement of the flux of the modified membrane.

In this context, the present study aims to evaluate the feasibility of the production of oil/water (O/W) emulsions by ME, using alumina-based monochannel hydrophilic ceramic membranes. The influence of process parameters on the droplet mean diameter, size distribution and emulsion stability was investigated. Two pore size membranes were tested (0.2 and 0.8 μm) and the sunflower oil was used as the disperse phase, due to its great applicability in the food industry.

2. Materials and methods

2.1. Reagents

Ultra-pure water (Milli-Q®) was used throughout the experimental runs. Sunflower oil (Sinhá, Paraná, Brazil) was purchased in the local market. The continuous phase was composed of Tween® 80 (polyethylene glycol sorbitan monooleate). Tween® 80 is a nonionic surfactant, with molecular mass of 1310 g mol^{-1} and critical micelle concentration (CMC) of 1.2×10^{-5} mol L^{-1}.

2.2. Membranes

Active-layer alumina-based ceramic membranes (Ceraver, France) were used. The membrane length was 0.25 m, the inner diameter 8×10^{-3} m and the external diameter 1×10^{-2} m. Therefore, the active membrane surface was 5×10^{-3} m^2. Two microfiltration membranes (mean pore size 0.2 μm and 0.8 μm) were investigated. According to the manufacturer, these membranes have a high porosity (55%) and high dispersion of pore size (0.1–0.35 μm for 0.2 μm-membrane, and 0.4–1.5 μm for 0.8 μm-membrane).

2.3. Experimental setup and assay procedure

The tests were carried out in a microfiltration system operating in cross flow mode, according Fig. 1. The display included a pump (PD 5025, Heidolph Instruments GmbH, Germany), a microfiltration membrane module and a stainless steel feed tank

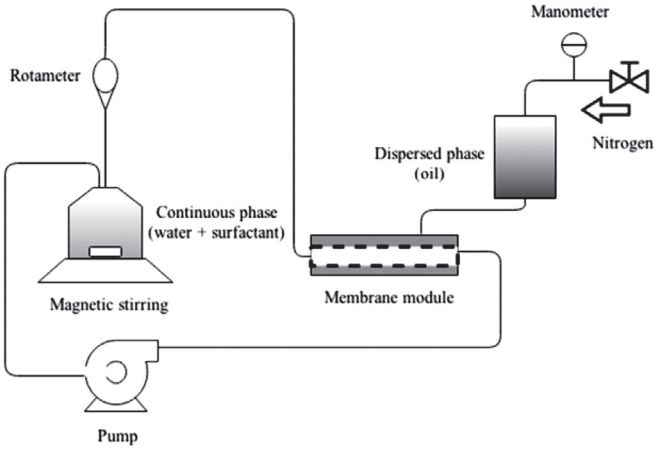

Fig. 1. Experimental setup for the preparation of O/W emulsions.

equipped with a manometer. The continuous phase, constituted of ultrapure water and surfactant, was continuously stirred (700 rpm) with a magnetic stirrer. A pressurized stainless steel vessel (300 mL), containing sunflower oil was equipped with a manometer, connected to a nitrogen bottle and to the membrane module on the filtrate side.

In all runs, membranes were immersed in aqueous phase for 12 h before emulsification run for pre-wetting. Prior to each experiment, the membranes were subjected to treatment with ultrasound for 30 min to eliminate possible air bubbles present in the pores. During the experiment, the continuous phase (ultrapure water containing Tween 80 as surfactant) was circulated tangentially through the membrane inner space. The dispersed phase (oil) was kept in the feed tank. The run was started ($t = 0$) opening the nitrogen valve, then pushing the disperse phase through the membrane pores, forming droplets that were emulsified into the aqueous phase. The experiment was stopped when all the desired amount of disperse phase was fed to the system. The emulsification process was run at 23 ± 2 °C for both membranes, as well as for all the subsequent characterizations.

At the end of the run, the microfiltration membrane was regenerated by flushing the experimental set-up in an open loop configuration with an aqueous solution of an alkaline detergent (1% v/v) (pH 10.5, VETEC) and ultrapure water. After washing, membranes were immersed in 0.1 M sodium hydroxide solution pH 12 for 20 min, followed by immersion for 20 min in 0.01% phosphoric acid solution pH 2. In the sequence, membranes were rinsed in ultrapure water, in which they were stored.

2.4. Experimental design

2.4.1. Membrane emulsification (ME)

This study was carried out using a Plackett & Burman design (Montgomery, 2005), comprised of eight linear experimental runs (PB8) with coded levels (−1 and +1) and three runs with central values (0), totalizing 11 experimental runs.

The following parameters were studied: pressure (kPa), tangential velocity ($m \cdot s^{-1}$), oil and surfactant concentration (%), and their levels are shown in Table 1.

2.4.2. Conventional emulsification (CE)

Conventional emulsions were prepared for comparison purposes with ME. These emulsions were prepared in a homogenizer Ultra digital Turrax® (model T 25, IKA®). The procedure consisted of stirring the continuous phase (water/surfactant) for 15 min to