



## Emulsification using tubular metallic membranes



Miguel A. Suárez, Gemma Gutiérrez, María Matos, José Coca, Carmen Pazos\*

Department of Chemical and Environmental Engineering, University of Oviedo, Julián Clavería, 8, 33006 Oviedo, Spain

### ARTICLE INFO

#### Article history:

Received 18 September 2013  
Received in revised form 11 April 2014  
Accepted 13 April 2014  
Available online 14 May 2014

#### Keywords:

Membrane emulsification  
Dimensionless numbers  
Oil-in-water emulsion  
Tubular metallic membrane  
Droplet formation regime

### ABSTRACT

Tubular metallic membranes with pore diameters of 5 and 10  $\mu\text{m}$  have been used in a cross-flow unit to prepare monodisperse oil-in-water emulsions (O/W) with span values as low as 0.67, significantly lower than for emulsions prepared with a rotor–stator homogenizer. The influence of typical operating parameters (continuous phase flow rate and transmembrane pressure) on droplet size distribution was studied. The smallest droplets were obtained at low transmembrane pressures and high continuous phase flow rates. The droplet production with tubular metallic membranes was higher than with other types of tubular membranes, such as SPG or ceramic.

Experimental results were compared with those obtained in a stirred tank unit operating under similar conditions and using flat metallic membranes with the same pore diameter. Droplet size prediction models based on simple force balances were applied to compare theoretical and experimental droplet diameters. The droplet formation regime (dripping, jetting) was also studied for both types of membranes.

© 2014 Elsevier B.V. All rights reserved.

### 1. Introduction

Emulsions are dispersions of two immiscible liquids which are commonly found in the pharmaceutical, food, chemical and cosmetic industries. Emulsions with a uniform and easy to control size are of great interest in some industrial applications, e.g. (i) preparation of water-in-oil (W/O) and oil-in-water (O/W) emulsions in the food industry, (ii) manufacture of monodisperse particles (tonners, chromatographic packings, etc.), and (iii) production of multiple emulsions and microcarriers (microcapsules, liposomes, niosomes) in the pharmaceutical industry.

A high shear stress is applied into the liquid mixture with conventional techniques such as high pressure homogenizers or rotor–stator devices containing the emulsion components. Elongation and cavitation play a more significant role in emulsification with ultrasounds. These techniques yield polydisperse emulsions, which are not suitable for some applications when a narrow droplet size distribution is required, such as in drug delivery systems.

Membrane emulsification has been used at a laboratory and pilot-plant scale to produce monodisperse emulsions since its introduction in the early 1990s [1–10] and is usually carried out by two techniques [11–15]: (i) *direct emulsification*, where one phase (the dispersed phase) is pushed through the membrane pores into a second phase (the continuous phase), and (ii) *premix*

*emulsification* [16], where an emulsion of coarse droplets is pushed through the membrane pores to produce the desired uniform emulsion. Microengineered devices (microchannels) also generate monodisperse droplets [17], and in some cases their operating principles are similar to membrane emulsification [18].

Membranes used for emulsification are manufactured from several materials, such as metals [19–25], organic polymers [26–31], and ceramics [5,6,10,32–35]. In addition, a special material called *Shirasu porous glass* (SPG) has been extensively used for direct membrane emulsification, yielding emulsions with very narrow droplet size distributions [1–4].

Highly monodisperse emulsions can be obtained with ceramic membranes but their performance depends on the control of the dispersed phase flux and the pore tortuosity. Frequently, drops are formed by a spontaneous mechanism [36–39], which has also been observed in emulsification with microchannels [40,41]. The main disadvantage of this mechanism is the low throughput [3,11,12].

Metallic membranes, which have been also used for emulsion microfiltration (MF) [42], have higher mechanical strength and longer operating life than ceramic membranes. Droplets are formed by a shear-based mechanism and the droplet size can be easily controlled by changing the operating parameters [20,43]. They also provide higher dispersed phase fluxes [25,44].

Some membrane emulsification work has been carried out with flat metallic membranes in stirred tanks [20–22,43,45] and cross-flow devices [46,47], while tubular metallic membranes have been used in rotating [24,25,48,49] and oscillating devices [50].

\* Corresponding author. Tel.: +34 985103509; fax: +34 985103434.  
E-mail address: [cpazos@uniovi.es](mailto:cpazos@uniovi.es) (C. Pazos).

## Nomenclature

$c_1$	correlation parameter in Eq. (12)
$c_2$	correlation parameter in Eq. (12)
$Ca$	capillary number
$Ca_\tau$	capillary number based on shear
$D_{10}$	diameter for which 10% of distribution volume has smaller size ( $\mu\text{m}$ )
$D_{50}$	diameter for which 50% of distribution volume has smaller size ( $\mu\text{m}$ )
$D_{90}$	diameter for which 90% of distribution volume has smaller size ( $\mu\text{m}$ )
$D_d$	mean droplet diameter ( $\mu\text{m}$ )
$D_d^*$	dimensionless droplet diameter: ratio between droplet and pore diameter ( $D_d/D_p$ )
$D_i$	impeller diameter (m)
$D_m$	tubular membrane internal diameter ( $\mu\text{m}$ ). Flat membrane active-area diameter (m)
$D_p$	membrane pore diameter ( $\mu\text{m}$ )
$D_T$	tank diameter (m)
$Eu$	Euler number
$H$	height of liquid (continuous phase) in the tank (m)
$J_d$	dispersed phase flux ( $\text{m}^3/\text{m}^2 \text{ s}$ )
$k_x$	wall correction factor for a simple sphere touching an impermeable wall
$L_m$	length of the membrane active area (m)
$n$	refractive index
$N$	rotational speed (rps)
$Oh_d$	dispersed phase Ohnesorge number
$P_d$	dispersed phase pressure (Pa)
$P_0$	continuous phase pressure (Pa)
$Q_c$	continuous phase flow rate ( $\text{m}^3/\text{s}$ )
$Q_{dp}$	dispersed phase flow rate through a single pore ( $\text{m}^3/\text{s}$ )
$t$	time (s)
$v_c$	continuous phase velocity (m/s)
$We_d$	dispersed phase Weber number
$We_{dtr}$	dispersed phase Weber number in the dripping to jetting transition
$Z$	distance from impeller to membrane surface (m)
<b>Greek letters</b>	
$\alpha_m$	exponent in Eq. (12)
$\Delta P$	transmembrane pressure (Pa)
$\Delta P_0$	continuous phase pressure drop in the tubular membrane (Pa)
$\mu_c$	continuous phase dynamic viscosity (Pa s)
$\mu_d$	dispersed phase dynamic viscosity (Pa s)
$\rho_c$	continuous phase density ( $\text{kg}/\text{m}^3$ )
$\rho_d$	dispersed phase density ( $\text{kg}/\text{m}^3$ )
$\sigma$	interfacial tension (N/m)
$\tau$	shear stress (Pa)

Membrane emulsification with tubular membranes in cross-flow operation has been extensively studied, mostly employing ceramic membranes [5–7,34,38,51] or SPG membranes [3,8,9,52,53]. However, little work has been reported on emulsification with tubular metallic membranes in cross-flow operation and employing large pore size membranes [19].

In a previous work [45], emulsification with flat metallic membranes in a stirred tank has been reported, at constant impeller tip speed. A correlation based on dimensional analysis was developed including the capillary and Euler numbers, to describe the process performance [43]. It was observed that the droplet formation using

MF ceramic membranes is a spontaneous process and cannot be described by a shear-based mechanism [39].

In the present work the performance of tubular metallic membranes in cross-flow equipment, using small pore sizes (5 and 10  $\mu\text{m}$ ), is reported. The effect of continuous phase flow rate and transmembrane pressure on droplet size distribution was investigated. Results were compared with calculated values from droplet size estimation models. Emulsions were also prepared using flat metallic membranes in a stirred tank, under similar operating conditions. Results were analyzed in terms of dimensionless numbers and droplet formation regimes and compared with those obtained with the tubular metallic membranes.

## 2. Materials and methods

### 2.1. Materials

Emulsions were prepared with a food-grade extra virgin olive oil ( $\mu_d = 51 \text{ mPa s}$ ,  $\rho_d = 886 \text{ kg}/\text{m}^3$ , and  $n = 1.4677$ , at 25 °C; acid value lower than 0.8) as the dispersed phase. They were stabilized with a non-ionic surfactant added to the aqueous phase, namely Tween 20® at 2 wt%, along with medium viscosity sodium carboxymethylcellulose with a degree of polymerization 1100 (CMCNa), at 0.01 wt% (both supplied by Sigma–Aldrich, USA).

Tailor-made nickel membranes (Micropore Technologies, Derbyshire, UK) with pore sizes of 5 and 10  $\mu\text{m}$  were used. These membranes had a regular pore array with 200  $\mu\text{m}$  of distance between pores, and 200  $\mu\text{m}$  of membrane thickness. These characteristics were identical both for tubular and flat membranes. Tubular membranes had a total length of 0.25 m (an active length of 0.20 m) and they were made of nickel. The nickel sheet was rolled and welded, yielding a cylindrical surface supported by two stainless steel ends. Their external diameter was 0.01 m. Flat metallic membranes (Micropore Technologies, Derbyshire, UK) had a diameter of 0.0470 m (active diameter of 0.0315 m), as previously reported [45].

### 2.2. Methods

#### 2.2.1. Emulsion characterization

A long bench Mastersizer S (Malvern instruments Ltd., UK) unit was used to measure droplet size distributions. Triplicate samples were prepared to determine the droplet size distribution for each emulsion. Mean droplet diameter ( $D_d$ ) corresponds to  $D_{50}$ , the standard percentile value from the analysis for which 50% of distribution volume has smaller size ( $\mu\text{m}$ ).

Dispersed phase content and flux were determined by measuring the dry oil matter of the emulsion. Five emulsion samples were collected at certain time intervals. Each triplicate sample was weighed into a porcelain crucible, and dried at 110 °C for 24 h. The crucible was cooled in a desiccator to room temperature under vacuum and weighed.

#### 2.2.2. Membrane emulsification experiments

All experiments were conducted at room temperature and replicated at least twice, being the standard deviation lower than 5  $\mu\text{m}$ . The coefficient of variation was in the ranges 0.2–5.0% and 0.7–5.2% for tubular and flat membranes, respectively.

**2.2.2.1. Cross-flow tubular unit.** The cross-flow equipment used in the emulsification experiments with tubular membranes is shown in Fig. 1 and it is similar to units using SPG [1,3,52,54] and ceramic [5,10,32] membranes, and it has been previously described for ultrafiltration (UF) experiments [55–59]. It was designed as pilot scale equipment and operated in recirculation mode of the emulsion to increase the volume fraction of dispersed phase.

Download English Version:

<https://daneshyari.com/en/article/686939>

Download Persian Version:

<https://daneshyari.com/article/686939>

[Daneshyari.com](https://daneshyari.com)