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Journal of Membrane Science 261 (2005) 136–144
journal of MEMBRANE SCIENCE
www.elsevier.com/locate/memsci

# Analysis of droplet size during crossflow membrane emulsification using stationary and vibrating micromachined silicon nitride membranes

Jie Zhu, David Barrow *

*Laboratory for Applied Microsystems, School of Engineering, Cardiff University, 1-5 The Parade, Cardiff CF24 3AA, UK*

Received 1 July 2004; received in revised form 1 December 2004; accepted 21 February 2005
Available online 21 April 2005

**Abstract**

Crossflow membrane emulsification is a promising method to achieve very small and uniform emulsions. The droplet size produced is controlled mainly by the choice of membrane. Using microengineering technology it is currently possible to produce membranes with precision defined parameters (uniform pore size, shape and inter-pore distance). In the work presented here, individual pore behaviour was studied using micromachined membranes with wider inter-pore distances (100 μm). It was found that the diameter of droplets increased during an initial period of operation. Also, interaction between droplets formed at adjacent pores was observed to enhance the reduction of mean droplet size and negatively correlated with inter-pore distance. A 'push-to-detach' mechanism was proposed to explain the behaviour observed. It was demonstrated that a micromachined membrane with pore diameter of 2 μm and inter-pore distance of 20 μm produced smaller droplets than for membranes with larger inter-pore distances. To facilitate the droplet detachment from the membrane and provide additional control over droplet detachment, the effects of membrane vibration were investigated. Preliminary results showed that smaller droplets could be produced by introducing low frequency (0–100 Hz) membrane vibrations without increasing their size distribution.


*Keywords:* Emulsification; Piezoactuated membrane; Micromac hined membrane

## 1. Introduction

Crossflow membrane emulsification (CME) was originally developed in Japan in the late 1980s [1]. Two main advantages of CME are identified [2]. First, the energy consumption per unit of product made using CME is much less than that using conventional methods, i.e., high-pressure homogeniser, the diaphragm homogeniser and the more recent microfluidizer. This is significant not only in term of energy efficiency but also in improving the quality and functionality of delicate ingredients since the high shear and accompanying temperature rise due to the viscous dissipation in conventional methods (99.8% energy converted to heat) have negative effects on the delicate ingredients. Secondly, CME is a much more promising method to achieve very small (especially diameter less than 1 μm) and uniform droplets than the conventional methods since the resulting droplet size is controlled not by the generation of turbulent droplet break-up, but primarily by the choice of membrane.

The most frequently used membranes as described in the literature are microporous glass (MPG) and Shirasu porous glass (SPG) [3]. These membranes are characterised by cylindrical, interconnected micropores. The ceramic α-$Al_2O_3$ or α-$Al_2O_3$ coated with titanium oxide or zirconia had also been used. The pore size can be reduced and pore size distribution made narrower by coating. These membranes come with different nominal pore size (0.05–14 μm) and distribution (usually claimed to be ±15%). It is generally accepted that the droplet diameter produced by CME can be linearly related to the pore size under given operating conditions:

$$d_d = x d_p$$

where $x$ can range typically from 2 to 10 [3–5] under optimised operating conditions and $d_d$, $d_p$ are the mean droplet diameter of emulsions and the pore diameter of membrane, re-

* Corresponding author. Tel.: +44 29 2087 5921; fax: +44 29 2087 4716.
*E-mail address:* barrow@cf.ac.uk (D. Barrow).

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doi:10.1016/j.memsci.2005.02.038

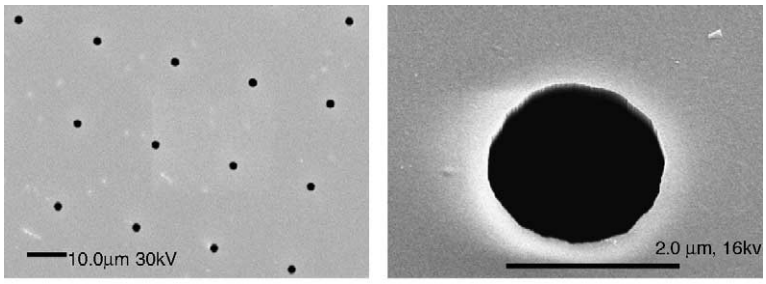


Fig. 1. Scanning electron micrograph of (a) a section of a micromachined silicon nitride membrane showing 14 pores and (b) detail of an individual pore.

spectively. Therefore, 'mono-dispersed' emulsions can only be produced if the membrane pore size distribution is sufficiently narrow. The presence of coarse pores can lead to bimodal distribution [6]. It is believed that the availability of well-defined membranes (uniform pore size, shape and inter-pore distance) could provide a step-change improvement in CME. Microengineering technology now enables the fabrication of such well-defined membranes in various formats. In this study, such micromachined membranes were used for investigation. An example of such a membrane (pore diameter 2.0 μm and inter-pore distance 20 μm) is shown in Fig. 1.

For a given system (continuous phase, dispersed phase, emulsifier and membrane are fixed), the most significant driving force for droplet detachment in CME is the shear force (or drag force) produced by continuous phase flow [4,7,12]. However, this provides only limited control over the process. It has been shown that membrane vibration can enhance local shear stress over the membrane [8]. Therefore, in the work reported here, membrane vibration, through piezoactuation, was investigated to help the detachment of droplets and provide an extra control over the emulsification process.

During this study, the individual pore behaviour in the emulsification process was visualised and the droplets generated were quantitatively analysed whilst using a specially designed membrane with relatively large inter-pore distance (the distance between adjacent pores). The observed pore behaviour provided valuable information for optimising experimental conditions and selecting membrane design. Thereafter, the performance of micromachined membranes with smaller inter-pore distance and pore size was also investigated. The investigation demonstrated that whilst significant further optimisation is required, vibrating micromachined membranes offer significant potential for CME.

## 2. Experimental

### 2.1. Materials

Micromachined (using both reactive ion etching and wet etching) silicon nitride membranes (surface area: 4 mm × 4 mm; thickness: 1.0 μm, Fig. 2) were provided by Aquamarijn Microfiltration B.V. (Netherlands). The specifications of these membranes are shown in Table 1.

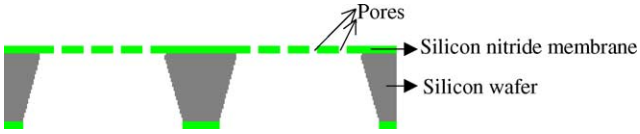


Fig. 2. A membrane pore array (drawing is two rows of pores).

Tween 20 (polyoxyethylene sorbitan monolaurate, Aldrich) dissolved in demineralized water was used as the continuous phase (CP). Hexadecane (BDH) coloured with saturated Sudan Red 7B (Aldrich) was used as the dispersed phase (DP). Prior to experiments, the DP was filtered using Polydisc$^{TM}$ PTFE membrane (Whatman) that has an average pore size of 0.45 μm. The CP was directly used without further treatment.

### 2.2. Experimental setup

The experimental setup is shown in Fig. 3. A flat membrane (Fig. 2) was housed in the module. The flow-rate of the CP was monitored by a water flow meter (Key Instruments; range 5–50 mL/min). The height of CP solution column was kept constant, which maintained a constant pressure over the membrane and a stable flow-rate in the channel of the module. A large container filled with aqueous solution was used to provide the continuous phase, without interruption over long periods (days). This design enabled life-time experiments on the membranes to be undertaken. The fluid channel for the CP in the module was 7.8 mm wide and 0.5 mm high. The dispersed phase was delivered by a KD Scientific (model 101) syringe pump, which had a minimum step of 1.0 μL/min with a 10 mL syringe. The formation of emulsions was observed and recorded by videomicroscopy using a custom variable

Table 1
Morphological specifications of the 1.0 μm thick micromachined silicon nitride membranes

| Code | Pore diameter (μm) | Inter-pore distance (μm) | Porosity (%) |
|---|---|---|---|
| M2.5L | 2.5 | 100 | 0.05 |
| M2.5 | 2.5 | 25 | 0.79 |
| M2.0 | 2.0 | 20 | 0.79 |