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Effects of crossflow velocity and transmembrane pressure on microfiltration of oil-in-water emulsions

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ABSTRACT

This study addresses the issue of oil removal from water using hydrophilic porous membranes. The effective separation of oil-in-water dispersions involves high flux of water through the membrane and, at the same time, high rejection rate of the oil phase. The effects of transmembrane pressure and crossflow velocity on rejection of oil droplets and thin oil films by pores of different cross-section are investigated numerically by solving the Navier–Stokes equation. We found that in the absence of crossflow, the critical transmembrane pressure, which is required for the oil droplet entry into a circular pore of a given surface hydrophilicity, agrees well with analytical predictions based on the Young–Laplace equation. With increasing crossflow velocity, the shape of the oil droplet is strongly deformed near the pore entrance and the critical pressure of permeation increases. We determined numerically the phase diagram for the droplet rejection, permeation, and breakup depending on the transmembrane pressure and shear rate. Finally, an analytical expression for the critical pressure in terms of geometric parameters of the pore cross-section is validated via numerical simulations for a continuous oil film on elliptical and rectangular pores.

1. Introduction

With the recent advances in environmental and biological technologies, there has been increasing interest in characterization and modeling flows at the micron scales including flows in microchannels and nanochannels [1,2], multiphase flows through porous media [3,4], and droplet-based microfluidics [5,6]. The industrial applications include oil extraction from porous media [7–9], treatment of oily wastewater [10–12], and encapsulation of molecules, cells, and microorganisms [13–15]. In most of these processes, one or more phases are dispersed in a continuous phase in the form of emulsions, which are usually produced by shearing two immiscible phases against each other in the presence of surfactants [16]. In some cases, emulsions serve as means of transport of molecules, bio-reagents, and drugs, and ultimately provide the environment for enhanced reactions [14,17]. Another technological application of emulsions is to improve the transportability or displacement of highly viscous liquids. For example, heavy crude oil is emulsified to form a less viscous mixture to facilitate its transportation [18,19]. In addition, oil-in-water emulsions are used to enhance recovery and increase sweep efficiency from crude oil reservoirs by blocking highly permeable paths and preventing channeling of the displacing fluid [9]. Common methods for separation of emulsions include evaporation of the continuous phase [20], destruction (demulsification) [21], and membrane filtration [10].

Membrane microfiltration has proven to be an efficient way for separating oil-in-water emulsions [22,23]. In comparison with the conventional methods of filtration (gravity separators, centrifuges, etc.), membrane microfiltration has several distinct advantages including reduced space requirements, higher permeate quality, and lower operating costs [10]. Despite its advantages, microfiltration efficiency can be greatly reduced because of membrane fouling at highly concentrated emulsions or long filtration times [24]. Fouling is generally caused by the accumulation of the rejected phase on the surface of the membrane or inside the pore. There are four main mechanisms (blocking laws) for membrane fouling, i.e., standard blocking, complete blocking, intermediate blocking, and cake formation [25]. Complete blocking is common for very dilute mixtures and during the initial stages of filtration when some pores are sealed by droplets and particles, thus reducing the permeate flux [26,27]. Accumulation of the rejected droplets on the membrane surface results in the formation of the so-called cake layer, which is sometimes referred to as the secondary membrane as it adds a hydraulic resistance to the microfiltration process [28–30]. This mechanism is dominant at the final stages of filtration when the water flux depends mainly on the thickness of the cake layer.

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The efficiency of the microfiltration process is determined by the properties of the membrane material and oil-in-water mixtures. For example, the permeate flux is highly dependent on the oil concentration, stability of the oil phase in water, and the size distribution of oil droplets [10,31]. Moreover, the membrane properties such as membrane material, pore size and morphology, and membrane geometry affect the permeate flow resistance [10,32]. It was shown that slotted (rectangular) pores resulted in higher flux rates compared to circular pores for similar operating conditions because of the lower fouling rate of the slotted-pore membranes [33]. Another approach to reduce the fouling rate is to introduce crossflow above the membrane surface. This method, known as the "crossflow microfiltration", reduces fouling by sweeping away the deposited drops and particles and, hence, decreases the thickness of the cake layer. Therefore, crossflow microfiltration systems tend to produce higher permeate fluxes for longer times compared to dead-end microfiltration systems [34,35]. One of the aims of the present study is to investigate numerically the entry dynamics of oil droplets into a membrane pore in the presence of crossflow.

The dynamics of droplet breakup in steady shear flow is determined by the relative competition of the viscous stress, pressure, and interfacial tension [36]. In general, the breakup process is initiated by the droplet deformation, which is linearly proportional to the rate of shear [37]. When the critical deformation is reached, the droplet assumes an unstable configuration and undergoes a transient elongation before it breaks up [36]. It was also shown that the geometric confinement as well as the viscosity ratio of the dispersed and continuous phases influence droplet breakup [38]. In recent years, the problem of droplet deformation and breakup has been extensively studied numerically using Lattice Boltzmann [39–41], boundary integral [42,43], and Volume of Fluid (VOF) [44–46] methods. The VOF method used in the present study has proven to be a powerful and efficient interface tracking algorithm that is both conceptually simple and relatively accurate [47]. Due to the conservative discretization of the governing equations in the VOF method, the mass of each fluid is accurately conserved [44,48]. Furthermore, the ability of the VOF method to automatically capture local and global changes of the interface topology, e.g., coalescence and breakup of droplets, has made it attractive for various multiphase flow applications [47].

During the last decade, a number of studies have investigated the process of droplet formation using cross-flowing streams in T-shaped junctions [40,49–51]. In these microfluidic systems, two immiscible liquids are driven through separate channels until their streams meet at a junction, where the dispersed liquid extends into the continuous stream, resulting in periodic formation of equal-sized droplets [52]. Regardless of the specific channel geometry and wettability of the channel walls, breakup of the emerging droplet in a cross-flowing stream is determined by the viscous drag when the droplet remains unconfined by the microchannel [52]. For unconfined T-junctions, it was demonstrated experimentally that the droplet size strongly depends on the crossflow rate of the continuous phase and only weakly on the flow rate of the dispersed phase [49,50]. It was also shown that, for a given value of continuous phase flow rate, the size of oil droplets decreases with increasing viscosity ratio of the oil and water [50].

In this paper, numerical simulations based on the Volume of Fluid method are performed to study the influence of transmembrane pressure and crossflow velocity on the entry dynamics of thin oil films and droplets into pores of various cross-sections. We find that the formula derived in Ref. [53] for the critical pressure of permeation of an oil droplet into a circular pore agrees well with the results of numerical simulations. The numerical analysis is then extended to thin oil films covering pores with elliptical and rectangular cross-section in the absence of crossflow. In the presence of crossflow, we obtain numerically the phase diagram for the droplet rejection, permeation, and breakup as a function of the transmembrane pressure and shear rate, and study the details of the processes in three different regions of the phase diagram. These results are relevant to microfiltration of dilute oil-in-water emulsions at early stages before the formation of the cake layer.

The rest of the paper is organized as follows. The details of numerical simulations are described in the next section. The analytical predictions based on the Young-Laplace equation are reviewed in Section 3.1 and verified numerically for an oil droplet on a circular pore in Section 3.2. The critical pressure of permeation for pores with elliptical and rectangular cross-section is reported in Section 3.3. The results for the oil droplet dynamics near circular pores in the presence of crossflow are presented in Section 3.4. The conclusions are given in the last section.

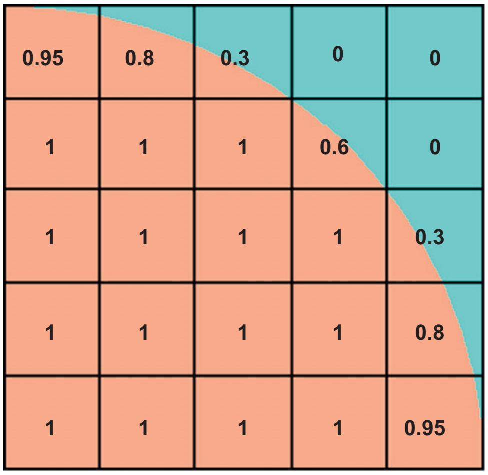
2. Details of numerical simulations

Numerical simulations were carried out using the commercial software FLUENT [54]. In order to control the transmembrane pressure and the crossflow velocity, a user-defined function was written and compiled along with the main solver. The Volume of Fluid method was used to solve the multiphase flow problem [55]. For a two-phase fluid, this method is based on the fact that the two phases form an impenetrable interface, i.e., each cell is filled with either one of the phases (denoting a specific phase zone) or a combination of two phases (denoting the interface). This is achieved by introducing a variable α, known as the "volume fraction", which is defined as the ratio of the volume of fluid in the cell and the total cell volume; and it varies between 0 and 1 [56,57]. An example of how the volume fraction varies near the interface is illustrated schematically in Fig. 1.

The interface is tracked by solving the transport equation for the volume fraction as follows:

$$\frac{\partial \alpha}{\partial t} + \nabla \cdot (\alpha \mathbf{V}) = 0, \tag{1}$$

where **V** is the velocity vector. Eq. (1) states that the substantial derivative of the volume fraction is zero, and, therefore, the interface is convected by the velocity field at the interface. After solving Eq. (1), the material properties are computed by considering the fraction of each component in the cell; e.g., the



0.95	0.8	0.3	0	0
1	1	1	0.6	0
1	1	1	1	0.3
1	1	1	1	0.8
1	1	1	1	0.95

Fig. 1. Values of the volume fraction for each of the phases and the interface ($\alpha = 1$ in the first phase, $\alpha = 0$ in the second phase, and $0 < \alpha < 1$ at the interface).