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Oil droplet behavior at a pore entrance in the presence of crossflow: Implications for microfiltration of oil–water dispersions

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ABSTRACT

The behavior of an oil droplet pinned at the entrance of a micropore and subject to crossflow-induced shear is investigated numerically by solving the Navier–Stokes equation. We found that in the absence of crossflow, the critical transmembrane pressure required to force the droplet into the pore is in excellent agreement with a theoretical prediction based on the Young–Laplace equation. With increasing shear rate, the critical pressure of permeation increases, and at sufficiently high shear rates the oil droplet breaks up into two segments. The results of numerical simulations indicate that droplet breakup at the pore entrance is facilitated at lower values of the surface tension coefficient, higher oil-to-water viscosity ratio and larger droplet size but is insensitive to the value of the contact angle. Using simple force and torque balance arguments, an estimate for the increase in critical pressure due to crossflow and the breakup capillary number is obtained and validated for different viscosity ratios, surface tension coefficients, contact angles, and drop-to-pore size ratios.

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1. Introduction

Understanding the dynamics of an oil droplet at a pore entrance is a fascinating problem at the intersection of fluid mechanics and interface science that is of importance in such natural and engineering processes as extraction of oil from bedrock, lubrication, aquifer smearing by non-aqueous phase liquids, and sealing of plant leaf stomata [1–4]. Membrane-based separation of liquid–liquid dispersions and emulsions is a salient example of a technology where the knowledge of liquid droplet behavior in the vicinity of a surface pore is critical for the success of practical applications. Milk fractionation, produced water treatment, and recovery of electrodeposition paint are examples of specific processes used in food, petroleum, and automotive industries where porous membranes are relied on to separate emulsions [5–7].

The membrane separation technique can be particularly useful when small droplets need to be removed from liquid–liquid dispersions or emulsions because other commonly used technologies, such as hydrocyclones and centrifugation-based systems, are either incapable of removing droplets smaller than a certain critical size (e.g., ~20 μm for hydrocyclones) or are expensive and have insufficient throughput (e.g., centrifuges). The early work by the Wiesner group [8] as well as other studies [9,10] on oil droplet

entry into a pore provided an estimate of the critical pressure of permeation; however, the understanding of the entire process of the droplet dynamics at a micropore entrance is still lacking, especially with regard to the practically relevant case of crossflow systems where blocking filtration laws [11] are, strictly speaking, not applicable. Crossflow membrane microfiltration is used to separate emulsions by shearing droplets of the dispersed phase away from the membrane surface and letting the continuous phase pass through [12]. In contrast to the normal, or dead-end, mode of filtration, crossflow microfiltration allows for higher permeate fluxes due to better fouling control [13,14]. However, the accumulation of the dispersed phase on the surface of the membrane and inside the pores, i.e., fouling of the membrane, can eventually reduce efficiency of the process to an unacceptably low level even in the presence of crossflow.

Another important application that entails interaction of liquid droplets with porous media is membrane emulsification, where micron-sized droplets are produced by forcing a liquid stream through membrane pores into a channel where another liquid is flowing [15]. The emerging droplets break when the viscous forces exerted by crossflow above the membrane surface are larger than surface tension forces [16]. Membrane emulsification requires less energy and produces a more narrow droplet size distribution [17,18] than conventional methods such as ultrasound emulsification [19] and stirring vessels [20].

In general, the studies of petroleum emulsions have been performed at two different scales, namely, macroscopic or bulk

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scales and mesoscopic or droplet scales [21,22]. Early research on membrane emulsification and microfiltration involved bulk experiments aimed at determining averaged quantities and formulating empirical relations [23]. These studies considered macroscopic parameters such as droplet size distribution, dispersed phase concentration, and bulk properties such as permeate flux [24,25]. These empirical approaches were adopted due to inherent complexity of two-phase systems produced by bulk emulsification, where shear stresses are spatially inhomogeneous and the size distribution of droplets is typically very broad [21,26]. However, with the development of imaging techniques and numerical methods, the shape of individual droplets during deformation and breakup could be more precisely quantified for various flow types and material properties [27,28].

First studies of the droplet dynamics date back to 1930s, when G.I. Taylor systematically investigated the deformation and breakup of a single droplet in a shear flow [29,30]. Since then, many groups have examined this problem theoretically [31,32] and in experiments [33–35]. A number of research groups have studied experimentally how a droplet pinned at the entrance of an unconfined pore deforms when it is exposed to a shear flow [17]. Experiments have also been performed to measure the size of a droplet after breakup as a function of shear rate and viscosity ratio [36,37]. Numerical simulations of the droplet deformation and breakup have been carried out using various methods including boundary integral [38], Lattice Boltzmann [39], and Finite Volume [40] methods. These multiphase flow simulations generally use an interface-capturing method to track the fluid interfaces. Among other front-tracking methods, the Volume of Fluid method simply defines the fluid–fluid interface through a volume fraction function, which is updated based on the velocity field obtained through the solution of the Navier–Stokes equation [41,42]. The Volume of Fluid method is mass-preserving, it is easily extendable to three dimensions, and it does not require special treatment to capture topological changes [43].

The drag force and torque on droplets or particles attached to a solid substrate and subject to flow-induced shear stress depend on their shape and the shear rate. Originally, O'Neill derived an exact solution for the Stokes flow over a spherical particle on a solid surface [44]. Later, Price computed the drag force on a hemispherical bump on a solid surface under linear shear flow [45]. Subsequently, Pozrikidis extended Price's work to study the case of a spherical bump with an arbitrary angle using the boundary integral method [46]. More recently, Sugiyama and Sbragaglia [47] varied the viscosity ratio to include values other than infinity (the only value considered by Price [45]) and found an exact solution for the flow over a hemispherical droplet attached to a solid surface. Assuming that the droplet is pinned to the surface, estimates for the drag force, torque, and the deformation angle as functions of the viscosity ratio were obtained analytically [47]. Also, Dimitrakopoulos showed that the deformation and orientation of droplets attached to solid surfaces under linear shear flow depend on the contact angle, viscosity ratio, and contact angle hysteresis [48].

More recently, Darvishzadeh and Priezjev [49] studied numerically the entry dynamics of nonwetting oil droplets into circular pores as a function of the transmembrane pressure and crossflow velocity. It was demonstrated that in the presence of crossflow above the membrane surface, the oil droplets can be either rejected by the membrane, permeate into a pore, or break up at the pore entrance. In particular, it was found that the critical pressure of permeation increases monotonically with increasing shear rate, indicating optimal operating conditions for the enhanced microfiltration process. However, the numerical simulations were performed only for one specific set of parameters, namely, viscosity ratio, contact angle, surface tension coefficient, and droplet-to-pore size ratio. One of the goals of the present

study is to investigate the droplet dynamics in a wide range of material parameters and shear rates.

In this paper, we examine the influence of physicochemical parameters such as surface tension, oil-to-water viscosity ratio, droplet size, and contact angle on the critical pressure of permeation of an oil droplet into a membrane pore. In the absence of crossflow, our numerical simulations confirm analytical predictions for the critical pressure of permeation based on the Young–Laplace equation. We find that when the crossflow is present above the membrane surface, the critical pressure increases, and the droplet deforms and eventually breaks up when the shear rate is sufficiently high. Analytical predictions for the breakup capillary number and the increase in critical permeation pressure due to crossflow are compared with the results of numerical simulations based on the Volume of Fluid method.

The rest of the paper is structured as follows. In the next section, the details of numerical simulations and a novel procedure for computing the critical pressure of permeation are described. In Section 3, the summary of analytical predictions for the critical pressure based on the Young–Laplace equation is presented, and the effects of confinement, viscosity ratio, surface tension, contact angle, and droplet size on the critical transmembrane pressure and breakup are studied. Conclusions are provided in the last section.

2. Details of numerical simulations

Three-dimensional numerical simulations were carried out using the commercial software ANSYS FLUENT [50]. The FLUENT flow solver utilizes a control volume approach, while the Volume of Fluid (VOF) method is implemented for the interface tracking in multiphase flows. In the VOF method, every computational cell contains a certain amount of each phase specified by the volume fraction. For two-phase flows, the volume fractions of 1 and 0 describe a computational cell occupied entirely by one of the phases, while any value in between corresponds to a cell that contains an interface between the two phases [51]. In our simulations, GAMBIT was employed to generate the mesh. In order to increase the simulation efficiency, we generated a hybrid mesh that consists of fine hexagonal meshes in a part of the channel that contains the droplet and coarse tetrahedral meshes in the rest of the channel. A user-defined function was used to initialize the droplet shape and to adjust the velocity of the top wall that induced shear flow in the channel, as shown schematically in Fig. 1.

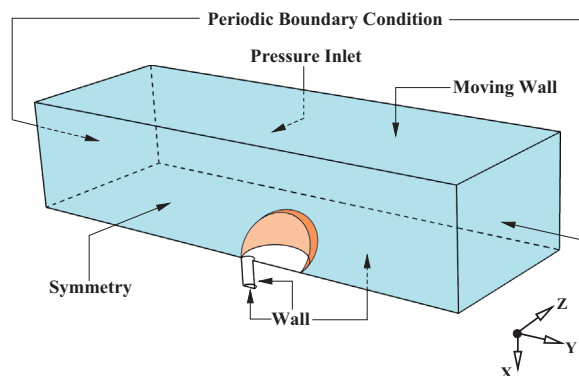


Fig. 1. Schematic representation of the oil droplet residing at the pore entrance in a rectangular channel with the corresponding boundary conditions. The width and length of the computational domain are fixed to $12\ \mu\text{m}$ and $36\ \mu\text{m}$, respectively. Symmetry boundary conditions are used in the z direction. The system dimensions are not drawn to scale.

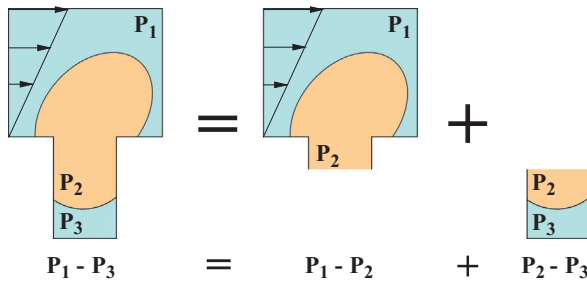


Fig. 2. Schematic of the droplet cross-sectional profile at the membrane pore. The critical pressure of permeation ($P_1 - P_3$) is calculated in three steps: (1) the pressure jump across the static interface ($P_2 - P_3$) is calculated from the Young–Laplace equation, (2) the pressure jump across the dynamic interface ($P_1 - P_2$) is computed numerically, and (3) the pressure jumps from steps 1 and 2 are added.

As we recently showed, the dynamics of the oil–water interface inside the pore slows down significantly when the transmembrane pressure becomes close to the critical pressure of permeation [49]. Hence, the interface inside the pore is nearly static and the pressure jump across the spherical interface is given by the Young–Laplace equation. However, numerical simulations are required to resolve accurately the velocity field, pressure, and shape of the deformed droplet above the pore entrance. In the present study, we propose a novel numerical procedure to compute the critical pressure of droplet permeation and breakup, as illustrated in Fig. 2. First, the pressure jump across the static interface inside the pore is calculated using the Young–Laplace equation. Second, we simulate the oil droplet in the presence of steady shear flow when the droplet covers the pore entrance completely and the oil phase partly fills the pore. In the computational setup, the pore exit is closed to prevent the mass flux and to keep the droplet at the pore entrance. The difference in pressure across the deformed oil–water interface with respect to the inlet pressure is measured in the oil phase at the bottom of the pore (see Fig. 2). The critical pressure of permeation is then found by adding the pressure differences from the first and second steps. In the previous study [49], the critical pressure of permeation at a given shear rate was determined iteratively by testing several transmembrane pressures close to the critical pressure. Using the novel approach, we were able to reproduce our previous results [49] faster and with higher accuracy. Moreover, this numerical procedure was automated to detect the critical pressure while increasing shear rate quasi-steadily, so that less post-processing is required.

The solution of the Navier–Stokes equations for the flow over the membrane surface requires specification of the appropriate boundary conditions. As shown in Fig. 1, there are four types of boundary conditions used in the computational domain. The membrane surface is modeled as a no-slip boundary. A moving “wall” boundary condition is applied at the top surface of the channel to induce shear flow between the moving top wall and the stationary membrane surface. The bottom of the pore is also described by the “wall” boundary condition to prevent the mass flux and to keep the oil droplet pinned at the pore entrance. Periodic boundary conditions are imposed at the upstream and downstream entries of the channel. On the lateral side of the channel in the ($Z+$) direction, a pressure-inlet boundary condition is applied to allow mass transfer, and to ensure that the reference pressure is fixed. Finally, a “symmetry” condition is implemented and only half of the computational domain is simulated to reduce computational efforts. We performed test simulations with an oil droplet $r_d = 2 \mu\text{m}$ exposed to shear flow and found that the local velocity profiles at the upstream, downstream, and the lateral sides remained linear when the width and length of the computational domain were fixed to $12 \mu\text{m}$ and $36 \mu\text{m}$,

respectively. These values were used throughout the study. The effect of confinement in the direction normal to the membrane surface on the droplet deformation and breakup will be investigated separately in Section 3.2.

The interface between two phases is described by a scalar variable, known as the volume fraction α , which is convected by the flow at every iteration via the solution of the transport equation as follows:

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

where \mathbf{V} is the three-dimensional velocity vector. The time dependence of the volume fraction is determined by the velocity field near the interface. Next, since the cells containing the interface include both phases, the material properties are averaged in each cell; for instance, the volume-fraction-averaged density is computed as follows:

$$\rho = \alpha \rho_2 + (1 - \alpha) \rho_1. \quad (2)$$

Using the averaged values of viscosity and density, the following momentum equation is solved:

$$\frac{\partial}{\partial t} (\rho \mathbf{V}) + \nabla \cdot (\rho \mathbf{V} \mathbf{V}) = -\nabla p + \nabla \cdot [\mu (\nabla \mathbf{V} + \nabla \mathbf{V}^T)] + \rho \mathbf{g} + \mathbf{F}, \quad (3)$$

where \mathbf{V} is the velocity vector shared between two phases, \mathbf{g} is the gravitational acceleration, and \mathbf{F} is the surface tension force per unit volume, which is given by

$$\mathbf{F} = \sigma \frac{\rho \kappa \nabla \alpha}{\frac{1}{2} (\rho_1 + \rho_2)}, \quad (4)$$

where σ is the surface tension coefficient and κ is the curvature of the oil–water interface, which in turn is defined as

$$\kappa = \frac{1}{|\mathbf{n}|} \left[\left(\frac{\mathbf{n}}{|\mathbf{n}|} \cdot \nabla \right) |\mathbf{n}| - (\nabla \cdot \mathbf{n}) \right], \quad (5)$$

where \mathbf{n} is the vector normal to the interface. The surface tension force given by Eq. (4) is nonzero only at the interface and it acts in the direction normal to the interface ($\mathbf{n} = \nabla \alpha$). Segments with higher interface curvature produce larger surface tension forces and tend to smooth out the interface [52]. The orientation of the interface at the wall is specified by the contact angle. The unit normal for a cell containing the interface at the wall is computed as follows:

$$\mathbf{n}_i = \mathbf{n}_w \cos \theta + \mathbf{n}_t \sin \theta, \quad (6)$$

where \mathbf{n}_w and \mathbf{n}_t are the unit vectors normal to the wall and normal to the contact line at the wall, respectively. The angle θ is the static contact angle measured in the dispersed phase [41].

A SIMPLE method was utilized for the pressure–velocity decoupling. A second order upwind scheme was used for discretization of the momentum equation and a staggered mesh with central differencing was used for the pressure equation. Piecewise Linear Interface Reconstruction (PLIC) algorithm was employed to reconstruct the interface in each cell [53]. The continuum surface force model of Brackbill et al. [41] was used to compute the surface tension force.

An accurate computation of the pressure and velocity fields for problems involving fluid interfaces requires a precise estimate of the interfacial curvature. It is well known that discrete formulation of an interface produces a loss of accuracy in regions of high curvature and, therefore, requires a sufficiently fine mesh. The numerical simulations were performed using the mesh size of $0.1 \mu\text{m}$, which corresponds to 32 mesh cells along the perimeter of the membrane pore. To ensure that the mesh resolution is sufficiently high, we performed simulations at different shear rates using 2 and 4 times finer meshes and found that the resulting refinements in the final position of the droplet interface

and the values of the critical permeation pressure were negligible. The total volume of the oil phase inside the pore and above the membrane surface was used to calculate the droplet radius. Unless otherwise specified, the following parameters were used throughout the study: the pore radius is $r_p = 0.5 \mu\text{m}$, the droplet radius is $r_d = 2 \mu\text{m}$, the contact angle measured in oil is $\theta = 135^\circ$, and the surface tension coefficient at the interface between water and oil is $\sigma = 19.1 \text{ mN/m}$.

3. Results

3.1. The critical pressure of permeation and the breakup capillary number

The pressure jump across a static interface between two immiscible fluids can be determined from the Young–Laplace equation as a product of the interfacial tension coefficient and the mean curvature of the interface or $\Delta P = 2\sigma\kappa$. For a pore of arbitrary cross-section, the mean curvature of the interface is given by

$$\kappa = \frac{C_p \cos \theta}{2A_p}, \quad (7)$$

where C_p and A_p are the cross-sectional circumference and area of the pore, respectively [54]. Therefore, the critical pressure of permeation of a liquid film into a pore of arbitrary cross-section is given by

$$P_{cr} = \frac{\sigma C_p \cos \theta}{A_p}. \quad (8)$$

In our recent study [49], the theoretical prediction for the critical permeation pressure, Eq. (8), was validated numerically for oil films on a membrane surface with rectangular, elliptical, and circular pores.

In the case of a liquid droplet blocking a membrane pore, the critical pressure of permeation, Eq. (8), has to be adjusted to account for the finite size of the droplet. It was previously shown [8,9] that the critical pressure for an oil droplet of radius r_d to enter a circular pore of radius r_p is given by

$$P_{cr} = \frac{2\sigma \cos \theta}{r_p} \sqrt[3]{1 - \frac{2 + 3 \cos \theta - \cos^3 \theta}{4(r_d/r_p)^3 \cos^3 \theta - (2 - 3 \sin \theta + \sin^3 \theta)}}. \quad (9)$$

We showed earlier that the analytical prediction for the critical pressure given by Eq. (9) agrees well with the results of numerical simulations for an oil droplet at the pore entrance in the absence of crossflow [49]. In the presence of crossflow, however, Eq. (9) is not valid as the shear flow deforms the droplet rendering its interface above the membrane surface non-spherical [49]. Furthermore, numerical simulations have shown that the critical pressure of permeation increases with increasing crossflow velocity up to a certain value, above which the droplet breaks up [49]. Hence, the phase diagram was determined for the droplet rejection, permeation, and breakup depending on the transmembrane pressure and shear rate [49]. In the present study, the critical permeation pressure is determined more accurately and its dependence on shear rate is studied numerically for a range of material properties and geometrical parameters.

In the presence of crossflow above the membrane surface, an oil droplet breaks up when viscous stresses over the droplet surface exposed to the flow become larger than capillary stresses at the interface of the droplet near the membrane pore. Therefore, at the moment of breakup, the drag force in the flow direction is balanced by the capillary force at the droplet interface around the

pore

$$D \approx F_\sigma. \quad (10)$$

Neglecting the contact angle dependence, $F_\sigma \propto \sigma r_p$ is the interfacial force acting in the direction opposite to the flow at the droplet interface near the pore entrance. The drag force generated by a linear shear flow on a spherical droplet attached to a solid surface is given by

$$D \propto f_D(\lambda) \mu \dot{\gamma} r_d^2, \quad (11)$$

where μ is the viscosity of the continuous phase, $\dot{\gamma}$ is the shear rate, and r_d is the radius of the droplet [47,55]. The coefficient $f_D(\lambda)$ is a function of the viscosity ratio $\lambda = \mu_o/\mu_w$ and it depends on the shape of the droplet above the surface. Sugiyama and Sbragaglia [47] have estimated this function analytically for a hemispherical droplet ($\theta = 90^\circ$) attached to a solid surface

$$f_D(\lambda) \approx \frac{2 + 4.510\lambda}{1 + 1.048\lambda}. \quad (12)$$

By plugging Eq. (11) into Eq. (10) and introducing $\bar{r} = r_d/r_p$, the critical capillary number for breakup of a droplet on a pore can be expressed as follows:

$$Ca_{cr} \propto \frac{1}{f_D(\lambda)\bar{r}}, \quad (13)$$

where the capillary number is defined as $Ca = \mu_w \dot{\gamma} r_d / \sigma$.

The difference in pressure inside the pore in the presence of flow and at zero shear rate can be estimated from the torque generated by the shear flow on the droplet surface. The torque around the center of the droplet projected on the membrane surface is given by

$$T \propto f_T(\lambda) \mu \dot{\gamma} r_d^3. \quad (14)$$

It was previously shown [47] that for a hemispherical droplet on a solid surface, $f_T(\lambda)$ is a function of the viscosity ratio

$$f_T(\lambda) \approx \frac{2.188\lambda}{1 + 0.896\lambda}. \quad (15)$$

Hence, the balance of the torque due to shear flow above the membrane surface [given by Eq. (14)] and the torque arising from the pressure difference, $(P_{cr} - P_{cr0})A_p r_d$, can be reformulated in terms of the capillary number and drop-to-pore size ratio as follows:

$$P_{cr} - P_{cr0} \propto \frac{f_T(\lambda) \sigma \bar{r} Ca}{r_p}, \quad (16)$$

where P_{cr0} is the critical permeation pressure in the absence of crossflow.

In what follows, we consider the effects of confinement, viscosity ratio, surface tension, contact angle, and droplet size on the critical pressure of permeation and breakup using numerical simulations and analytical predictions of Eqs. (13) and (16).

3.2. The effect of confinement on droplet deformation and breakup

In practical applications, the dimensions of a crossflow channel of a microfiltration system are much larger than the typical size of emulsion droplets so that the velocity profile over the distance of about r_d from the membrane surface can be approximated as linear. To more closely simulate this condition in our computational setup, the shear flow above the membrane surface was induced by moving the upper wall of the crossflow channel (Fig. 1). To understand how the finite size of the channel affects droplet dynamics at the membrane surface, we studied the influence of the channel height on the droplet behavior. The confinement ratio is defined as the ratio of the height of the droplet residing on the pore at zero shear rate H_d (i.e., the height of

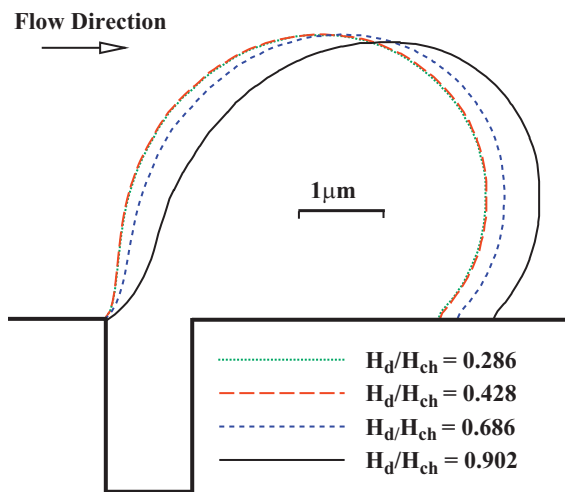


Fig. 3. The cross-sectional profiles of oil droplets in steady shear flow for the indicated confinement ratios when the capillary number is $Ca=0.021$. The droplet radius is $r_d = 2 \mu\text{m}$, the pore radius is $r_p = 0.5 \mu\text{m}$, the contact angle is $\theta = 135^\circ$, the surface tension coefficient is $\sigma = 19.1 \text{ mN/m}$, and the viscosity ratio is $\lambda = 1$.

a spherical cap above the membrane surface) to the channel height H_{ch} . It is important to note that the degree of confinement is varied only in the direction normal to the membrane surface and the computational domain is chosen to be wide enough for the lateral confinement effects to be negligible (see Section 2).

We performed numerical simulations of an oil droplet with radius $r_d = 2 \mu\text{m}$ in steady-state shear flow for the channel heights $3.8 \mu\text{m} \leq H_{ch} \leq 12.0 \mu\text{m}$. Fig. 3 illustrates the effect of confinement on the shape of the droplet residing on a $r_p = 0.5 \mu\text{m}$ pore when the capillary number is $Ca = \mu_w \dot{\gamma} r_d / \sigma = 0.021$. The height of the droplet above the membrane surface in the absence of flow is approximately $3.43 \mu\text{m}$. It can be observed from Fig. 3 that highly confined droplets become more elongated in the direction of flow than droplets with lower confinement ratios, which is in agreement with the results of previous simulations [56]. When a droplet is highly confined, the distance between the upper moving wall and the top of the droplet is relatively small. As a result, the effective shear rate at the surface of the droplet is higher and the droplet undergoes larger deformation. Furthermore, the cross-sectional profiles for the confinement ratios of 0.428 and 0.286 are nearly identical, indicating that the flow around the droplet is not affected by the upper wall when $H_d/H_{ch} \leq 0.428$ and the capillary number is fixed.

Fig. 4 shows the variation of the critical capillary number (right before breakup) as a function of the confinement ratio for the same parameters as in Fig. 3. These results indicate that highly confined droplets breakup at lower capillary numbers, and, when the confinement ratio is smaller than about 0.5, the breakup capillary number remains nearly constant. For the rest of the study, the channel height was fixed to $8 \mu\text{m}$, which corresponds to

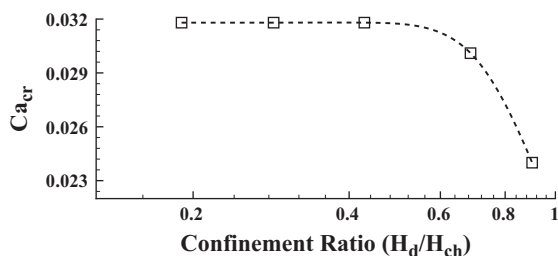


Fig. 4. The critical (breakup) capillary number as a function of the confinement ratio H_d/H_{ch} . Other parameters are the same as in Fig. 3.

the confinement ratio of 0.428 for a droplet with radius $r_d = 2 \mu\text{m}$. For the results presented in Section 3.6, the channel height was scaled appropriately to retain the same confinement ratio for larger droplets.

3.3. The effect of viscosity ratio on the critical transmembrane pressure

The ratio of viscosities of the dispersed and continuous phases is an important factor that determines the magnitude of viscous stresses at the interface between the two phases. For a small droplet at low Reynolds numbers, the viscous stresses are primarily counterbalanced by interfacial tension stresses. In a shear flow, viscous stresses tend to distort the surface of a droplet, while interfacial stresses assist in retaining its initial spherical shape. The competition between the two stresses determines the breakup criterion, deformation, and orientation of the droplet [28,57]. In this subsection, we investigate numerically the effect of viscosity ratio on the droplet deformation and breakup at the entrance of the membrane pore.

Fig. 5 shows the effect of the viscosity ratio, $\lambda = \mu_o/\mu_w$, on the critical pressure of permeation and breakup of an oil droplet on a membrane pore as a function of the capillary number. The percent increase in critical pressure is defined with respect to the critical pressure in the absence of crossflow P_{cr_0} , i.e., $(P_{cr} - P_{cr_0})/P_{cr_0} \times 100\%$. Keeping in mind that P_{cr_0} does not depend on λ , the results shown in Fig. 5 demonstrate that at a fixed Ca , the critical pressure increases with increasing viscosity ratio, which implies that higher viscosity droplets penetrate into the pore at higher transmembrane pressures. Specifically, the maximum increase in critical pressure just before breakup is about 8% for $\lambda = 1$ and about 15% for $\lambda = 20$. Furthermore, highly viscous droplets tend to break at lower shear rates because of the larger torque generated by the shear flow [see Eq. (14)]. As reported in Fig. 5, the critical capillary number for breakup varies from about 0.018 for $\lambda = 20$ to 0.032 for $\lambda = 1$. The practical implication of these results is that in membrane emulsification processes the use of liquids with lower viscosity ratios should be avoided as the droplets tend to break at higher shear rates.

Examples of cross-sectional profiles of the oil droplet in steady shear flow are presented in Fig. 6 for the viscosity ratio $\lambda = 1$. At

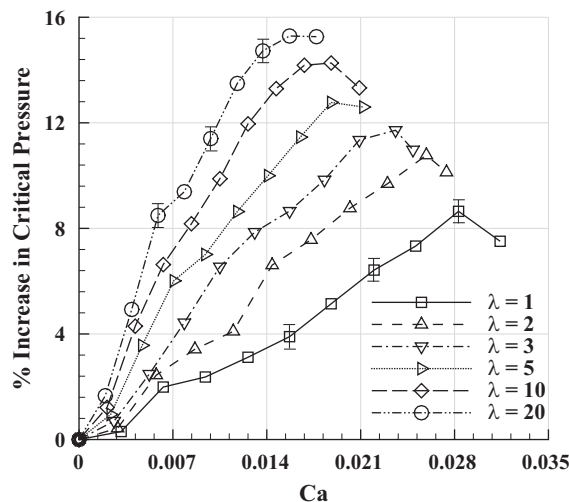


Fig. 5. The percent increase in critical pressure of permeation as a function of the capillary number $Ca = \mu_w \dot{\gamma} r_d / \sigma$ for the indicated viscosity ratios $\lambda = \mu_o/\mu_w$. Typical error bars are shown on selected data points. For each value of λ , the data are reported up to the critical capillary number above which droplets break into two segments. The droplet and pore radii are $r_d = 2 \mu\text{m}$ and $r_p = 0.5 \mu\text{m}$, respectively. The contact angle is $\theta = 135^\circ$ and the surface tension coefficient is $\sigma = 19.1 \text{ mN/m}$.

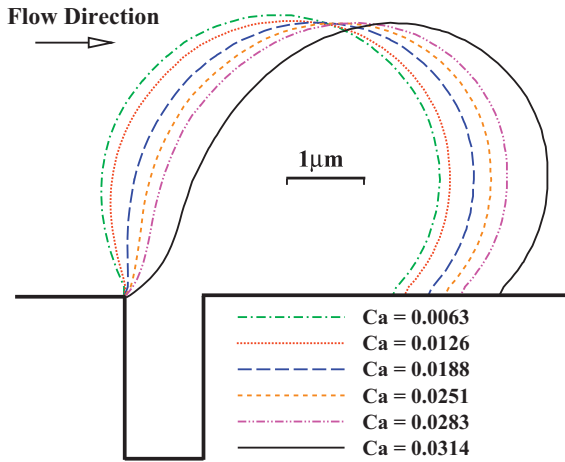


Fig. 6. The cross-sectional profiles of the oil droplet residing on the circular pore with $r_p = 0.5 \mu\text{m}$ for the indicated capillary numbers. The viscosity ratio is $\lambda = 1$. Other parameters are the same as in Fig. 5.

small capillary numbers, no significant deformation occurs and the droplet retains its spherical shape above the membrane surface. As Ca increases, a neck forms at the pore entrance while the rest of the droplet remains nearly spherical. A closer look at the shapes of the droplet for $Ca = 0.0283$ and 0.0314 in Fig. 6 reveals that with increasing shear flow, the neck gets thinner and the droplet becomes more elongated in the direction of flow. While the torque due to the shear flow does not increase significantly, the elongated shape of the droplet results in an effectively longer arm for the torque due to pressure in the droplet along the flow direction, and, thus, it leads to a lower critical permeation pressure required to keep the droplet attached to the pore. This effect is observed in Fig. 5 as the critical pressure just before breakup decreases as a function of Ca .

We next estimate the breakup time and compare it with the typical deformation time of the droplet interface for different viscosity ratios. In our simulations, the upper wall velocity is increased quasi-steadily and the spontaneous initiation of the breakup process can be clearly detected by visual inspection of the droplet interface near the pore entrance. We then identify the moment when a droplet breaks into two segments and compute

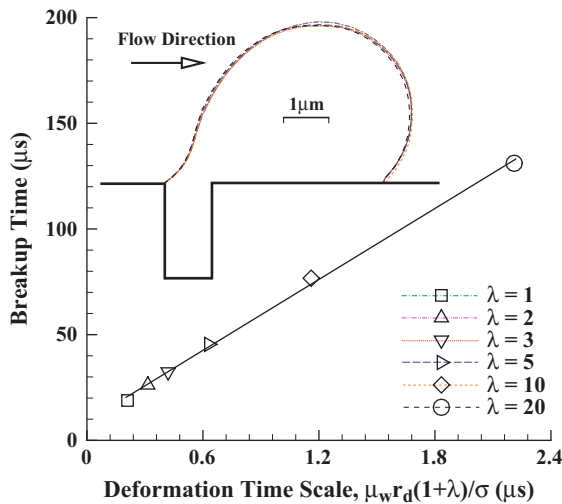


Fig. 7. The breakup time versus deformation time scale $\mu_w r_d (1 + \lambda) / \sigma$ for the tabulated values of the viscosity ratio $\lambda = \mu_o / \mu_w$. Other system parameters are the same as in Fig. 5. The straight line is the best fit to the data. The error bars for the breakup time are about the symbol size. The inset shows the droplet profiles just before breakup for the same viscosity ratios.

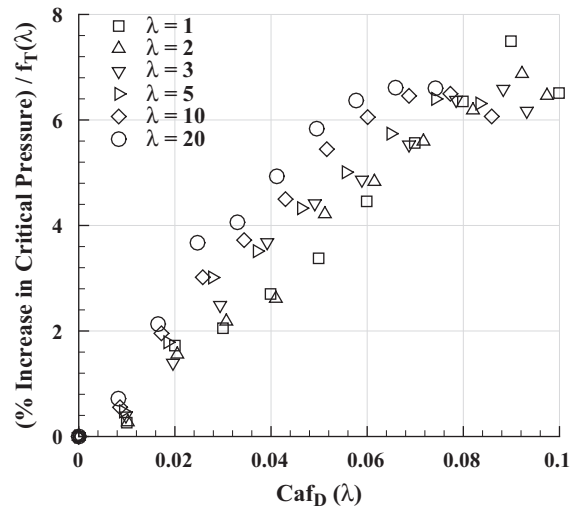


Fig. 8. The normalized percent increase in critical pressure of permeation versus the modified capillary number $Ca f_D(\lambda)$ for the selected values of the viscosity ratio $\lambda = \mu_o / \mu_w$. The functions $f_D(\lambda)$ and $f_T(\lambda)$ are given by Eqs. (12) and (15), respectively.

the breakup time. The deformation time scale, defined by $\mu_w r_d (1 + \lambda) / \sigma$, is a measure of the typical relaxation time of the droplet interface with respect to its deformation at steady state [48,58]. In Fig. 7, the breakup time is plotted against the deformation time scale for different viscosity ratios. Notice that the breakup time increases linearly with the deformation time scale, which confirms that highly viscous droplets break up more slowly. The inset in Fig. 7 displays the droplet cross-sectional profiles just before breakup for the same viscosity ratios. It can be observed that the profiles nearly overlap with each other, indicating that droplets with different viscosities are deformed identically just before breakup.

According to Eq. (13), the breakup capillary number depends on the viscosity ratio via the function $f_D(\lambda)$. Therefore, it is expected that the product $Ca_{cr} f_D(\lambda)$ will be independent of λ and the appropriate dimensionless number for a constrained viscous droplet in a shear flow is $Ca f_D(\lambda)$. Moreover, based on Eq. (16), the percent increase in the critical pressure is independent of the viscosity ratio when it is divided by $f_T(\lambda)$. Fig. 8 shows the same data as in Fig. 5 but replotted in terms of the normalized critical pressure and the modified capillary number. As is evident from Fig. 8, the data for different viscosity ratios nearly collapse on the master curve. It is seen that droplets break at approximately the same value $Ca f_D(\lambda) \approx 0.09$. In practice, the increase in critical pressure due to crossflow can be roughly estimated from the master curve in Fig. 8 for any viscosity ratio in the range $1 \leq \lambda \leq 20$. Also, if $Ca f_D(\lambda) \geq 0.09$, the oil droplets will break near the pore entrance for any viscosity ratio.

3.4. The effect of surface tension on the critical pressure of permeation

In this subsection, we investigate the influence of surface tension on the critical permeation pressure, deformation and breakup of an oil droplet residing at the pore entrance in the presence of crossflow above the membrane surface. Fig. 9 shows the critical pressure of permeation as a function of shear rate for five values of the surface tension coefficient. As expected from Eq. (9), the critical pressure at zero shear rate increases linearly with increasing surface tension coefficient. Note that oil droplets with higher surface tension break up at higher shear rates because larger stresses are required to deform the interface and cause breakup of the neck. Also, the difference between the critical

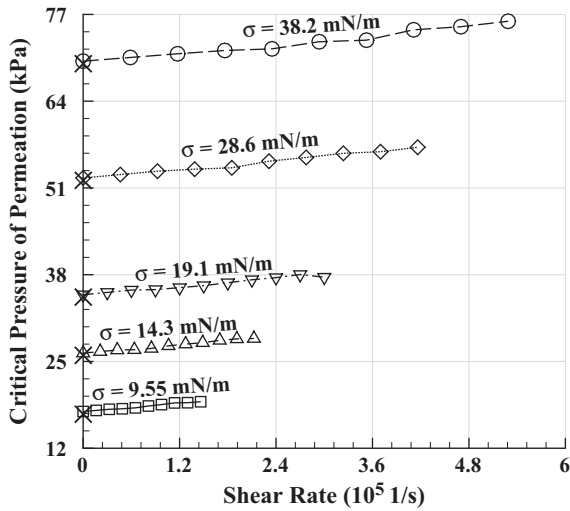


Fig. 9. The critical pressure of permeation as a function of shear rate for the indicated surface tension coefficients. The symbols (\times) denote the analytical predictions of Eq. (9). The droplet and pore radii are $r_d = 2 \mu\text{m}$ and $r_p = 0.5 \mu\text{m}$, respectively. The viscosity ratio is $\lambda = 1$ and the contact angle is $\theta = 135^\circ$.

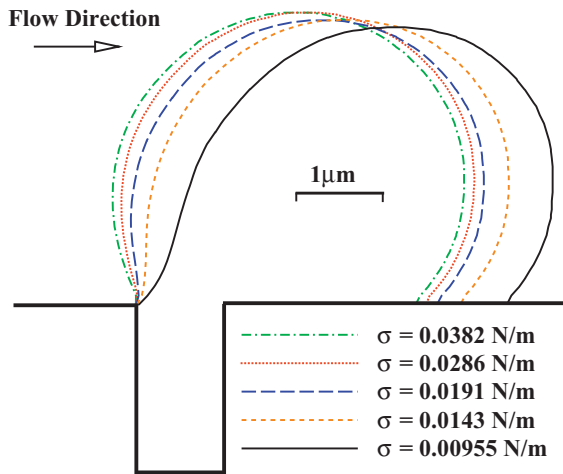


Fig. 10. The cross-sectional profiles of the oil droplet above the circular pore for the listed values of the surface tension coefficient. In all cases, the shear rate is $\dot{\gamma} = 1.5 \times 10^5 \text{ s}^{-1}$. Other parameters are the same as in Fig. 9.

pressure just before breakup and P_{cr0} is larger at a higher surface tension; for example, it is about 1.5 kPa for $\sigma = 9.55 \text{ mN/m}$ and 6 kPa for $\sigma = 38.2 \text{ mN/m}$. The results shown in Fig. 9 suggest that crossflow microfiltration of emulsion droplets with higher surface tension is more efficient because higher transmembrane pressure can be applied and the droplet breakup is less likely.

Examples of droplet cross-sectional profiles above the membrane pore are presented in Fig. 10 for five values of the surface tension coefficient. These profiles are extracted from the data reported in Fig. 9 at the shear rate $\dot{\gamma} = 1.5 \times 10^5 \text{ s}^{-1}$. It can be observed that oil droplets with lower surface tension become highly deformed along the flow direction. The elongation is especially pronounced when the surface tension coefficient is small; for $\sigma = 9.55 \text{ mN/m}$ the droplet interface is deformed locally near the pore entrance and the neck is formed.

To further investigate the effect of surface tension on the droplet breakup, we compare the breakup time and the deformation time scale $\mu_w r_d (1 + \lambda) / \sigma$. The numerical results are summarized in Fig. 11 for the same values of the surface tension coefficient as in Fig. 9. Similar to the analysis in the previous subsection, the breakup time was estimated from the time when a droplet

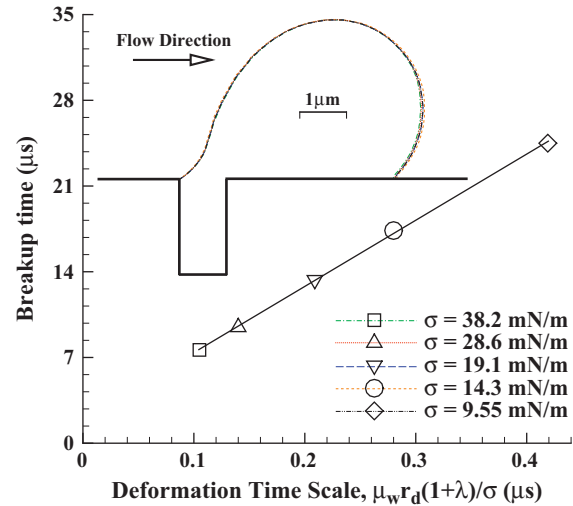


Fig. 11. The breakup time versus deformation time scale $\mu_w r_d (1 + \lambda) / \sigma$ for the surface tension coefficients in the range from 9.55 mN/m to 38.2 mN/m. Other parameters are the same as in Fig. 9. The straight solid line is the best fit to the data. The cross-sectional profiles of the oil droplet just before breakup are displayed in the inset.

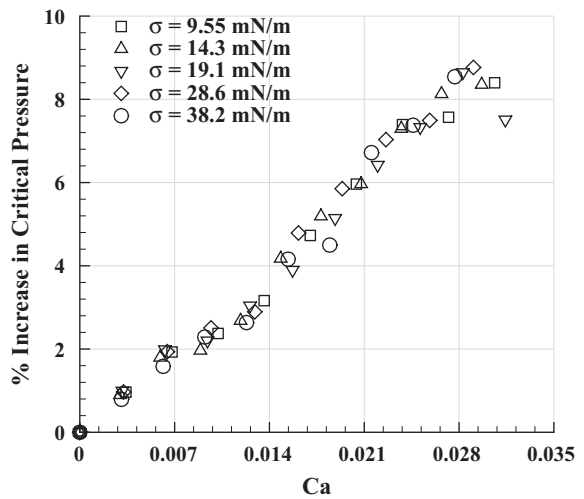


Fig. 12. The percent increase in critical pressure of permeation as a function of the capillary number $Ca = \mu_w \dot{\gamma} r_d / \sigma$ for the selected values of the surface tension coefficient. The rest of the parameters are the same as in Fig. 9.

becomes unstable under quasi-steady perturbation till the formation of two separate segments. It can be observed in Fig. 11 that the breakup time varies linearly with increasing deformation time scale, which in turn indicates that the breakup time is inversely proportional to the surface tension coefficient. In addition, the inset in Fig. 11 shows the cross-sectional profiles of the droplet just before breakup for the same surface tension coefficients. Interestingly, the profiles nearly coincide with each other, indicating that the droplet shape at the moment of breakup is the same for any surface tension.

In order to present our results in a more general form, we replotted the data from Fig. 9 in terms of the percent increase in critical pressure, $(P_{cr} - P_{cr0}) / P_{cr0} \times 100\%$, and the capillary number in Fig. 12. Note that in all cases, the data collapse onto a master curve and breakup occurs at the same relative pressure $(P_{cr} - P_{cr0}) / P_{cr0} \approx 8\%$ and $Ca_{cr} \approx 0.03$, which indicates that the capillary number is an appropriate dimensionless number to describe the droplet deformation in shear flow with variable surface tension. These results are not surprising, given that the breakup

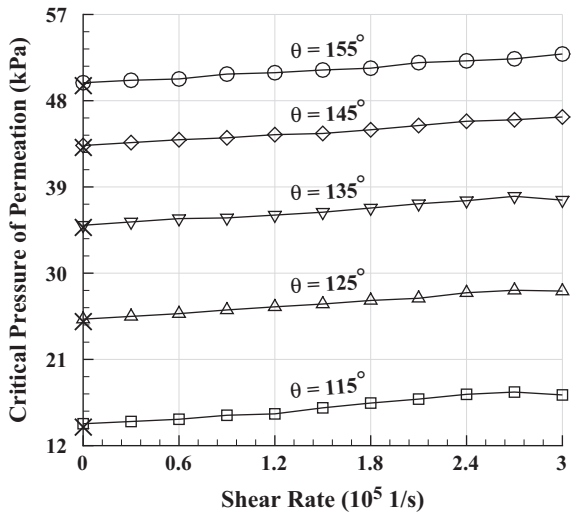


Fig. 13. The critical pressure of permeation as a function of the capillary number for the indicated contact angles. The critical pressure at zero shear rate, given by Eq. (9), is denoted by the symbols (\times). The droplet radius, pore radius, surface tension coefficient, and viscosity ratio are $r_d = 2 \mu\text{m}$, $r_p = 0.5 \mu\text{m}$, $\sigma = 19.1 \text{ mN/m}$, and $\lambda = 1$, respectively.

capillary number, Eq. (13), does not depend on the surface tension coefficient. Moreover, the increase in critical pressure due to crossflow, Eq. (16), is proportional to σ and Ca , and when it is divided by P_{cr0} , which itself is a linear function of σ [see Eq. (9)], the percent increase in critical pressure becomes proportional to the capillary number. In practice, the master curve reported in Fig. 12 can be used to predict the critical permeation pressure and breakup of emulsion droplets for specific operating conditions and surface tension.

3.5. The effect of contact angle on the droplet dynamics near the pore

Next, we focus on the effect of contact angle on the permeation pressure, deformation and breakup of oil droplets on a membrane pore. The variation of the critical permeation pressure as a function of the capillary number is presented in Fig. 13 for nonwetting oil droplets with contact angles $115^\circ \leq \theta \leq 155^\circ$. The critical pressure at zero shear rate is higher for oil droplets with larger contact angles, which is in agreement with the analytical prediction of Eq. (9). As expected, with increasing shear rate, the critical pressure of permeation increases for all values of θ studied. We estimate the maximum change in the critical pressure to be about 3 kPa and roughly independent of the contact angle. This corresponds to a relative increase of about 6% for the contact angle $\theta = 155^\circ$ and 21% for $\theta = 115^\circ$. These results suggest that the relative efficiency of a microfiltration system due to crossflow is higher for emulsion droplets with lower contact angles. Interestingly, we find that the critical capillary number for breakup ($Ca_{cr} \approx 0.032$) is nearly independent of the contact angle. This suggests that Ca can be used as a criterion for predicting breakup. Finally, the examples of the droplet cross-sectional profiles are shown in Fig. 14 for different contact angles when $Ca = 0.022$. Notice that droplets with lower contact angles wet larger solid area and are less tilted in the direction of flow.

3.6. The effect of droplet size on the critical pressure of permeation

In the microfiltration process, the size of the membrane pore is one of the crucial parameters that determine the permeate flux and membrane selectivity. Membranes with smaller pore sizes provide higher rejections but require higher transmembrane

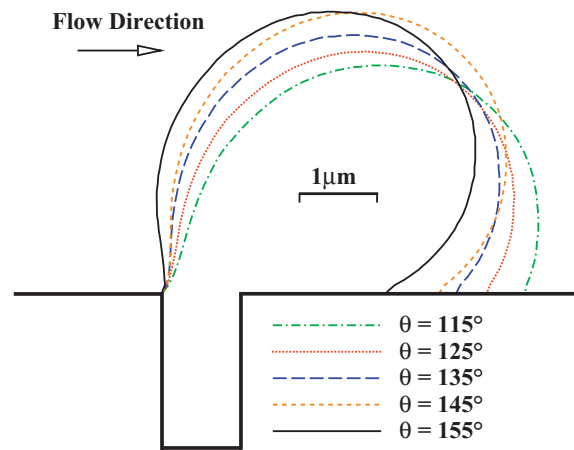


Fig. 14. The cross-sectional profiles of the oil droplet above the circular pore for the listed values of the contact angle when $Ca = 0.022$. Other parameters are the same as in Fig. 13.

pressures to achieve the same permeate flux. In this subsection, we examine the influence of the drop-to-pore size ratio on the critical pressure of permeation and the breakup dynamics of oil droplets in the presence of crossflow above the membrane surface.

Fig. 15 reports the critical permeation pressure as a function of shear rate for the droplet radii in the range from $1.5 \mu\text{m}$ to $2.5 \mu\text{m}$, while the pore radius is fixed at $r_p = 0.5 \mu\text{m}$. In the absence of crossflow, the critical pressure is higher for larger droplets because they have lower curvature of the interface above the membrane surface, which is in agreement with the analytical prediction of Eq. (9). With increasing shear rate, the critical pressure increases for droplets of all sizes. Note also that the slope of the curves in Fig. 15 is steeper for larger droplets because of the larger surface area exposed to shear flow, resulting in a higher drag torque, and, consecutively, a higher transmembrane pressure needed to balance the torque. Furthermore, as shown in Fig. 15, smaller droplets break at higher shear rates, since higher shear stress is required to produce sufficient deformation for the breakup to occur. The maximum relative critical pressure is about 14% for $r_d/r_p = 3$ and 6% for $r_d/r_p = 5$.

We next compute the difference in the critical permeation pressure with respect to the critical pressure in the absence of

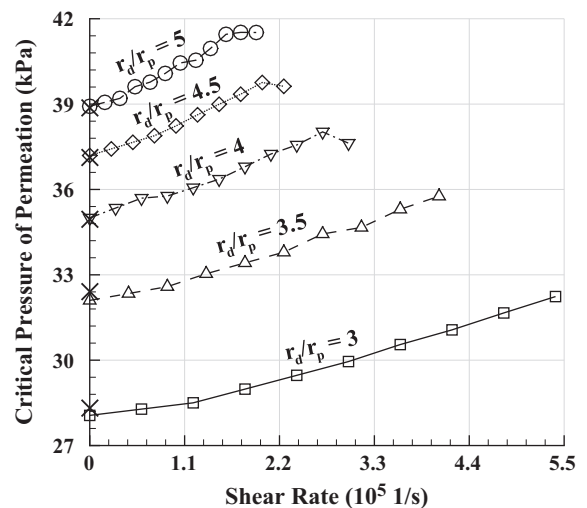


Fig. 15. The critical pressure of permeation as a function of shear rate for the selected drop-to-pore size ratios. The symbols (\times) indicate the critical pressure in the absence of flow calculated from Eq. (9). The pore radius, surface tension coefficient, contact angle, and viscosity ratio are $r_p = 0.5 \mu\text{m}$, $\sigma = 19.1 \text{ mN/m}$, $\theta = 135^\circ$ and $\lambda = 1$, respectively.

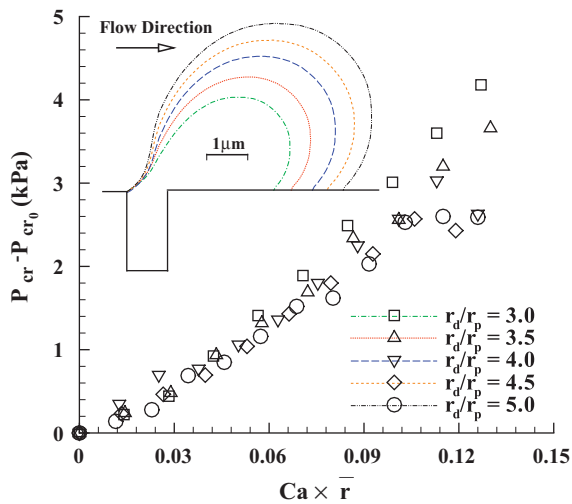


Fig. 16. The difference in the critical pressure, $P_{cr} - P_{cr0}$, versus the modified capillary number for five drop-to-pore size ratios $\bar{r} = r_d/r_p$ and $r_p = 0.5 \mu\text{m}$. Other parameters are the same as in Fig. 15. The cross-sectional profiles of the droplet just before breakup are shown in the inset for same \bar{r} .

flow, $P_{cr} - P_{cr0}$, and define $\bar{r} = r_d/r_p$. According to Eq. (13), the product $Ca_{cr} \times \bar{r}$ is independent of the droplet radius. At the same time, Eq. (16) suggests that the increase in critical pressure depends on the droplet radius via the term $Ca \times \bar{r}$. Fig. 16 shows the critical pressure difference as a function of the modified capillary number $Ca \times \bar{r}$ for different droplet radii. It can be observed in Fig. 16 that all curves nearly collapse on each other and the droplet breakup occurs at the same value $Ca \times \bar{r} \approx 0.125$. We also comment that one of the assumptions in deriving Eq. (16) is that the distance between the center of the pore and the center of the droplet on the membrane surface is approximately r_d . This approximation becomes more accurate for larger drop-to-pore size ratios, and, thus, the critical pressure difference in Fig. 16 is nearly the same for larger droplets even at high shear rates.

The inset of Fig. 16 shows the cross-sectional profiles of oil droplets just before breakup for different drop-to-pore size ratios. Note that all droplets are pinned at the pore entrance and elongated in the direction of flow. It is seen that when \bar{r} is small, the droplet shape is significantly deformed from its original spherical shape. In contrast, larger droplets remain nearly spherical and only deform near the pore entrance. In general, the droplet-to-pore size ratio should be large enough to make P_{cr} sufficiently high for practicable separation. At the same time, if the pore size is much smaller than the droplet size, the water flux through the membrane decreases and the probability of breakup increases, which could result in lower rejection rates and internal fouling of the membrane. Therefore, choosing a membrane with an appropriate pore size could greatly increase the efficiency of the microfiltration process.

4. Conclusions

In this paper, we performed numerical simulations to study the effect of material properties on the deformation, breakup, and critical pressure of permeation of oil droplets pinned at the membrane pore of circular cross-section. In our numerical setup, the oil droplet was exposed to a linear shear flow induced by the moving upper wall. We used finite-volume numerical simulations with the Volume of Fluid method to track the interface between water and oil. The critical pressure of permeation was computed using a novel procedure in which the critical permeation pressure was found by adding pressure jumps across oil–water interfaces of

the droplet inside the pore and above the membrane surface. First, the pressure jump across the static interface inside the pore was calculated using the Young–Laplace equation. Then, the pressure jump across the dynamic interface above the membrane surface was computed numerically and added to the pressure jump inside the pore. This method has proven to be accurate, robust, and computationally efficient. To determine the dimensions of the computational domain, we also studied the effect of confinement on the droplet deformation and breakup and concluded that in order to minimize finite size effects and computational costs, the distance between the membrane surface and the upper wall has to be at least twice the droplet diameter. In particular, it was observed that highly confined droplets become significantly deformed in a shear flow and break up more easily.

In the absence of crossflow, we found that the analytical prediction for the critical permeation pressure derived by Nazzal and Wiesner [8] agrees well with the results of numerical simulations for different oil-to-water viscosity ratios, surface tension coefficients, contact angles, and droplet sizes. In general, with increasing crossflow shear rate, the critical permeation pressure increases with respect to its zero-shear-rate value and the droplet undergoes elongation in the flow direction followed by breakup into two segments. The results of numerical simulations indicate that at a fixed shear rate, the critical permeation pressure increases as a function of the viscosity ratio, which implies that more viscous droplets penetrate into the pore at higher trans-membrane pressures. In agreement with a scaling relation for the critical capillary number, we also found that droplets of higher viscosity tend to break at lower shear rates. Furthermore, with increasing surface tension coefficient, the maximum increase in the critical permeation pressure due to crossflow becomes larger and the droplet breakup occurs at higher shear rates. Interestingly, the percent increase in critical permeation pressure as a function of the capillary number was found to be independent of the surface tension coefficient. Next, we showed that the breakup capillary number and the increase in critical pressure of permeation are nearly independent of the contact angle. Last, it was demonstrated that smaller droplets penetrate into the pore at lower pressures and break up at higher shear rates because larger shear stresses are needed to deform the interface above the membrane surface.

While most microfiltration membranes used in medium- to large-scale separation applications have pores of complex morphologies and a distribution of nominal sizes, results obtained for the simple case of a pore of circular cross-section can be useful for identifying general trends. As the model describes the interaction of oil droplets with unblocked pores, which corresponds to the initial stage of filtration or the stage that immediately follows membrane cleaning, the results can be helpful in understanding how fouling starts and in devising means to delay the onset of fouling. With the development of new methods of manufacturing micro-engineered membranes [59] and the rapid growth in the diversity and scale of applications of microfluidic devices, conclusions obtained in this work can be of direct practical value for guiding membrane design and optimizing process variables.

Acknowledgments

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