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The porous media's effect on the permeation of elastic (soft) particles

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ABSTRACT

To further the ability to design membranes for separation/fractionation of deformable particles (such as, cells, liposomes, vesicles, and droplets in emulsions and oil-water suspensions), we have developed a 2-d multiscale computational approach to study how the pressure drops and bulk flow within the depth of a porous "membrane" influences the mobility of an immiscible droplet through that structure. We use a combination of the extended finite element method to describe the creeping fluid flow ($Re \sim 0$) inside a portion of a filtration membrane with an embedded fluid droplet, coupled with a particle method that interpolates the droplet's interfacial position, as well as, the corresponding velocity and pressure fields using least square fitting. We calculated how the combination of several model 2-d porous network domain geometries (pore size and distribution), and a soft particle's deformation-related property (surface tension), influences the particles' velocity relative to the bulk fluid flux (aka sieving) in model porous domains made up of circular obstacles. The focus in this paper is on the scaling relationship between the particle's properties, the geometry of the system, and the overall droplet's sieving through a periodic domain. We present first the case of a droplet permeating through an individual pore to determine its critical pressure. Then, the base case of a single pore and droplet is extended to include arrays of obstacles (creating a porous network domain) with different droplet volume fraction. In this case, the applied trans-domain pressure gradient is not the same pressure drop each droplet experiences when it needs to deform in order to pass between obstacles (a pore throat), which results in a non-intuitive motion. These cases can provide a set of scaling rules to guide membrane design for droplet separation purposes.

1. Introduction

The transport of soft colloids, vesicles and other small deformable particles, such as cells, bacteria, immiscible droplets (oil-in-water and other emulsions [1]) and macromolecule aggregates, through a porous medium, is relevant to the development and use of filtration membranes for processes involved in applications related to food [2–4], water, life sciences [5], and energy [6]. Notably, in the pharmaceutical industry, the fabrication and purification of efficient drug carriers in the form of soft vesicles or liposomes [7–9] often rely on filtration processes, and studies of deformable particles' transport through pores have been conducted for years in the context of blood filtration [10,11], vesicles as drug carriers [12], simulations of biological membranes [13], as well as microfiltration (MF) of cells [14], and fluid sterilization [15]. In addition, microfluidic devices [16–18] developed to capture circulating tumor cells in the blood—for early diagnosis and better treatment solutions—rely on similar physical phenomena.

Filtration studies of deformable particles have often focused on how particle deformation within a depositing "cake" increases the resistance to flow by compression [19–22]. Recently, Darvishzadeh [23] modeled the critical pressure response of oil droplets, pinned at a pore entrance, to increasing shear rate and forecasted "break up" of the droplets, before pore penetration, with sufficient shear. As a follow-up, Tummons et al. [24] not only provided a comprehensive introduction and motivation for applications relevant to oil-in-water emulsions in the context of MF and ultrafiltration (UF) membrane processes, but also, showed experimental confirmation of the trends elucidated by Darvishzadeh [23] using direct microscopic observations. On the other hand, the transport of immiscible droplets and/or soft, deformable colloidal particles entering and exiting a membrane, is relevant to cases wherein a deposition cake may not be forming, also in oily-water filtrations [25]. Holdich et al. [26] experimentally identified that long, narrow, slotted pores better impede the passage of deformable droplets versus circular pores because flow passes around the droplet through the unobstructed portion of the slot-pore. Other work followed in the similar vein of slotted pore membranes [27–31], with Ullah et al. [29] determining the equilibrium deformation force required to reshape a spherical, surface droplet into a prolate ellipsoidal one that could enter the converging (non-parallel-edged) slotted surface pore, if subjected to sufficient drag force from the suspension's flux. This work was extended

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[30] to consider non-converging (parallel-edged) slotted pores and a droplet deforming into an oblate ellipsoid. The authors calculated that the deformation force required for pore entry would be greater for the non-converging case, thus further illustrating the importance of pore shape and flow redirection.

Earlier studies on emulsion (oil-in-water with surfactant) formation and fractionation, by Park et al. [32], investigated the operating pressure at which droplets could split up during their interaction with pores. They found that below a critical pressure, the pre-formed droplets could permeate pores by deforming into ellipsoids, but recovered their original shape upon exiting from the pore. Above the critical pressure however, the same droplets broke off into smaller droplets under the action of shear stress. This latter critical pressure was theoretically set as the capillary pressure, based on the diameter of the pore alone, and was experimentally found to be an accurate viewpoint. The phenomenon of droplet permeation could therefore be split into two regimes: one, at lower pressure gradients, in which the droplets deform to flow through the pore and a second, at higher pressure gradients, in which the original droplets break off into smaller size droplets.

It is important to note that in the majority of prior theoretical and experimental studies, an idealized membrane with straight-through pores, mostly symmetrical, has been used, with most of the focus being on droplet penetration through surface pores. By contrast, most real membranes have an interconnected, internal pore structure, that may include depth effects. For example, Boom et al., [33] discussed how the underlying, interconnected porous network of Shirasu Porous Glass membranes could explain why these membranes have been so effective at creating relatively uniform emulsion droplet distributions. They speculated that, because of pressure equilibration between the external (continuous) phase and the near-surface pore structure due to internal lateral flow, a "pinch-off" of dispersed phase droplets could occur at a more uniform size (given by the surface pore's Laplace pressure). This phenomenon is in contrast to the creation of larger drops arising from a continuous flow through a straight-through pore. Thus, not only for immiscible droplet sieving, but also membrane emulsification processes, it is meaningful to develop model-based, scaling rules for how deformable particles, will transport through an interconnected porous network domain. Furthermore, to the best of our knowledge there are no systematic studies relating the internal membrane structure to the kinetics of droplet(s) velocity through a porous medium—relative to the bulk fluid flux. This, of course, is the basis of the separation (or sieving) properties in a rate-based separation process.

As noted by Celia [34], there are different ways to model a multiphase flow in a porous media depending on the desired outcome of the model. Many approaches concentrated on finding macroscopic properties of the materials using multiscale approaches to the problem [35–37]. However, to fully understand the physics behind these processes, one must account in more detail for the effects at a microscopic length scale [38]. In this regard, several models account for the pore-fluid interactions to determine the effect of capillary phenomena on the behavior of such fluids [39,40]. These methods are however not suitable to model the physics of discrete particle systems. More recent approaches use Lattice-Boltzmann [41] approaches to simulate and study the dynamics of fluid droplets traveling in a porous network, which allows them to determine the overall behavior of the porous media, but tend to neglect the particle physics at the microscopic scale [42].

The aim of the current study is to illustrate an initial (and computationally tractable) approach for evaluating the individual effects of particle deformability, pore size, and network geometry on the droplet permeation (mobility) through porous media. We first study how a soft droplet permeates through different individual pores. We then address how this permeation is affected when the same nominal case scenario is part of a larger media domain, which results in a non-intuitive mobility. In this context, Foucard et al. [43] proposed a method to describe the transport of soft vesicles with an elastic interface through porous media at the microscale. Using that same approach, we have modeled the permeation of a soft particle through an idealized porous network (in 2-d), and determined how both the geometry of the porous media and the soft particle influence the particles' sieving relative to the bulk fluid flux (velocity). In this paper, we describe the derivation and non-dimensionalization of the model system; then, a thorough study of both fluid and deformable droplets in a pore is performed; and finally, we extend these results to a microscopic portion of a filtration membrane.

2. Visual motivation

To illustrate the phenomenon modeled in this work, we performed an experiment wherein a PDMS microfluidic channel was fabricated to observe immiscible droplets' deformation through a porous network. In this experiment, the porous network is idealized as an array of circular pillars arranged in a square array (pillar diameter and spacing are nominally 44 and 16 μm, respectively). The particles are water-in-oil droplets (~30 μm diameter) coated with SPAN-80 to stabilize them and prevent the droplets from merging. Further details of the experiment can be found in the Supplemental information.

From this experiment, we made two main observations whose physics will be addressed with the following numerical modeling. First, we observe that after the droplets experience a certain amount of deformation, they jump from one pore to the next, which is the onset of an unstable situation after a critical pressure/deformation is achieved [44,45]. Second, we observe that while the top particle, which is slightly larger in diameter, is trapped in the pore, the flow is able to circumvent that blockage and push the smaller two particles located below. Once these droplets have moved downstream, the pressure drop across the larger (upper) droplet becomes sufficient to facilitate its deformation and permeation through the pore. These images illustrate that the internal pressure (and flow) redistribution plays a critical role determining whether or not a particle can permeate through a porous network.

3. Permeation of an individual droplet through a single pore

The objective of this section is to quantitatively investigate the relationship between particle size and deformability, and its critical pressure; i.e. the pressure that enables its movement through a narrow pore formed by two circular obstacles. This problem, including different pore shapes and contact angles, was investigated in 3-d by the authors in [44], and only the essential details will be discussed in this article. We focus here on micron-sized soft particles such that (1) their interface can be modeled using surface tension and (2) they have a contact angle of 180° (non-wetting). Strictly speaking, the first assumption is only valid on fluid droplets. Nonetheless, the size of some lipid coated vesicles such as large liposomes or neutrophils allow to neglect bending effects and model their behavior using only surface tension [45–48]. Hence, in the following the term droplet is meant to include all these particle types. Similarly, the wetting of the obstacles (pores) by the particles would introduce a higher complexity in the model which would not enrich the physical insights herein discussed. It would only add an additional force by which the pore attracts the droplet [44]. The problem therefore degenerates to the study of an immiscible droplet squeezing through a narrow channel, for which numerical studies have traditionally been proposed, most notably within the context of micropipette aspiration [49].

In the present case, we consider the confined space between two circular 'obstacles' of radius b separated by a distance $2s$ (Fig. 2a). Let us further consider that an incompressible, non-wetting fluid vesicle with interfacial tension γ is trapped within this space as it is subjected to a pore pressure difference $\Delta P = P_1 - P_2$. From a theoretical viewpoint, the shape of this droplet can be divided into three parts that