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Micro-scale simulation of unidirectional capillary transport of wetting liquid through 3D fibrous porous media: Estimation of effective pore radii

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ABSTRACT

The effective and representative pore radii (R_{50}) determined from the physical experiments such as capillary penetration and porosimetry were found to be different for porous materials like ceramic and particulate structures. These differences were attributed to contact angle variations, possibly induced by contaminations. In this paper, we studied the relationship between effective and representative pore radii using "numerical experiments", in which the contact angle variations were eliminated by using a constant contact angle. Micro-scale simulations of capillary penetration of a wetting liquid (air–water configuration) through virtual fibrous media were performed using a finite-volume-based volume-of-fluid (VOF) method. From the simulations, it was found that unidirectional liquid penetration followed Lucas–Washburn kinetics ($L \sim \sqrt{t}$) at the micro-scale, except during the initial stages that are dominated by the inertial forces. Even though the contact angle was held constant in the numerical simulations, the effective pore radii of the fibrous structures differed significantly from the representative pore radii (R_{50}). The results suggest that a unique value for the characteristic pore radius does not exist for a porous medium, and the value of characteristic pore radius is strongly dependent on the experimental technique used to characterize the medium. Thus, the choice of experimental technique (e.g. porosimetry or capillary penetration experiment) to determine a characteristic pore radius should be based on the end application of the porous material.

Introduction

Fibrous porous materials are used in a wide variety of applications as they are soft, structurally flexible, and have relatively high resistance to mechanical deformation (Ashari and Vahedi Tafreshi, 2009; Pradhan et al., 2012; Soltani et al., 2014). Capillary penetration of a liquid through a fibrous porous medium is an important process that occurs in many applications, including, but not limited to, printing, drug delivery patches, textile industries, sanitary wipes, and performance fabrics (sports apparel) (Ashari and Vahedi Tafreshi, 2009; Hong and Kim, 2007; Hyvaluoma et al., 2006; Pradhan et al., 2012; Simile, 2004). Understanding the kinetics of capillary penetration of liquid is essential for the characterization of fibrous media, and for the optimization and development of new products (Jaganathan et al., 2008b).

Historically, unidirectional capillary transport of a liquid in porous media with a distinct liquid propagating front (i.e. saturated flow behind the wetting front) is described using the bundle-of-capillaries approach (Hyvaluoma et al., 2006; Lu et al., 2013; Zhmud et al., 2000). In this approach, a porous medium is assumed to consist of bundle of parallel capillary tubes of same radii, and the kinetics of liquid penetration through the medium is described by the Lucas–Washburn equation (Eq. 1).

$$L = \sqrt{\left(\frac{\sigma R_c cos\theta}{2\mu}\right)t}. \tag{1}$$

Here, L is the location of liquid propagating front, R_c is equivalent capillary radius of the porous medium, θ is the contact angle made by the wetting liquid on the solid fiber, t is the wetting time, σ is the surface tension between wetting and non-wetting phases, and μ is the dynamic viscosity of the wetting liquid. Derivation of the Lucas–Washburn equation is provided in Appendix A. The Lucas–Washburn equation has found applications in a variety of research areas, such as contact angle measurement, development of viscometers, and characterization of porous media (O'Loughlin et al., 2013; Xue et al., 2006). Additionally, a modified Lucas–Washburn equation (derivation is provided in Appendix B) that represents the viscous

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losses using the Darcy law has been proposed in the literature.

$$L=\sqrt{\left(\frac{4\sigma\, k\cos\theta}{\mu\varphi R_s}\right)t}, \tag{2}$$

where, R_s, k, and ϕ are static radius, permeability and porosity of the medium, respectively. It should be noted that the capillary penetration models described in Eqs. (1) and (2) are based on simplifying assumptions, and the applicability of these models to real porous media with tortuous pore space is still under investigation (Hyvaluoma et al., 2006; O'Loughlin et al., 2013).

Lucas–Washburn kinetics ($L\sim\sqrt{t}$): In the unidirectional capillary penetration experiments conducted at the macro-scale, the distance traveled by the wicking front or the height rise was found to be proportional to the square root of time (Fries and Quéré, 2010; Hyvaluoma et al., 2006; Masoodi, 2010). Typically, these experiments are carried out at length scales much larger than the structural inhomogeneities in a porous medium (Hyvaluoma et al., 2006). One of the interesting questions is to understand if the liquid imbibition at the micro-scale (length scales close to and even below those of structural variations) follows Lucas–Washburn kinetics ($L\sim\sqrt{t}$) (Bousfield and Karles, 2004; Gane et al., 2004; Ridgway and Gane, 2002). If the liquid imbibition follows Lucas–Washburn kinetics at all scales, then $L\sim\sqrt{t}$ behavior might be intrinsic to the propagation of meniscii in a capillary penetration process. In the present effort, liquid penetration kinetics at the micro-scale (fiber-level) was studied by simulating unidirectional capillary transport of a wetting liquid through 3D virtual fibrous structures using a finite-volume-based volume-of-fluid (VOF) method, which relies only on conservation principles and allows for explicit description of geometry and interface dynamics (Berberović et al., 2009; Deshpande et al., 2012; Ferrari and Lunati, 2013).

Characterization of porous media: In applications where the rate of capillary penetration is important, characterization of porous media usually involves determination of an effective pore radius either equivalent capillary radius (R_c) or static radius (R_s), and an effective contact angle. However, the effective pore radius and effective contact angle of fibrous media cannot be independently determined from the results of a single capillary penetration (or wicking) experiment (Lavi et al., 2008; Marmur, 2003). Typically, this is achieved by performing two experiments (two-fluid method). A reference liquid, assumed to form a zero contact angle with the fibrous medium, is used to calculate the effective pore radius, and then the effective contact angle is calculated from experiments with the liquid of interest (Lavi et al., 2008).

The pore space of fibrous media can also be characterized using porosimetry techniques, which give a distribution of pore-sizes in a fibrous network (Jaganathan et al., 2008a). A representative pore radius (R_{50}), the pore radius at which 50% of the non-wetting liquid intrusion volume occurs, is typically reported as it is considered a well-accepted representative radius for a micro-structure with a distribution of pore features (Schoelkopf et al., 2002). It should be noted that the approach of using representative pore radius (R_{50}) to characterize a porous medium with a distribution of pores appears to lack a solid scientific basis.

Even though the goal of the characterization effort is to determine an unique characteristic pore radius, the effective pore radii obtained from capillary penetration experiments were typically found to be different from the representative pore radii obtained from pore-size distribution data (Hanžič et al., 2010). Einset et al. (1996) studied liquid imbibition rates into particulate structures of carbon and silicon carbide, and found a discrepancy of 1–2 orders of magnitude between effective pore radius and representative pore radius. Li et al. (1994) conducted wicking experiments in ceramic structures using apolar, low-energy liquids, and reported an effective pore radius smaller than the representative pore radius by a factor of approximately two. Although there is no reason for the effective and representative pore radii of a porous medium to be identical from a physics point of view, a single-valued characteristic pore radius was assumed in literature (Einset et al., 1996; Hanžič et al., 2010; Li et al., 1994; Schoelkopf et al., 2002). The disparity between the values of effective and representative pore radii obtained from experiments were attributed to the variation in contact angle in experiments, possibly induced by contaminations, which to some extent are inevitable (Hamraoui and Nylander, 2002; Li et al., 1994; Schoelkopf et al., 2002).

In contrast to the physical experiments, detailed numerical simulations of capillary penetration of a wetting liquid through porous media can be performed by keeping the contact angle constant (thereby eliminating the effects of contact angle variations) to study the relationship between effective and representative pore radii. To the best of the authors' knowledge, no other study has been reported in the literature to numerically determine the effective pore radius of fibrous media. In the present effort, transient two-phase flow simulations of unidirectional capillary penetration of a wetting liquid (water) through 3D virtual fibrous structures media were performed with a constant contact angle using a finite-volume-based volume-of-fluid (VOF) method. Firstly, it will be investigated whether the liquid penetration through fibrous media follows Lucas–Washburn kinetics at the micro-scale. The penetration rates are then used to determine effective pore radii of the fibrous structures. The effective pore radii calculated from the micro-scale simulations are compared with the representative pore radii estimated from the pore-size distribution data.

Methodology

In the first part of this section, the construction of virtual 3D fibrous structures and determination of pore-size distributions are described. The flow conservation equations and the corresponding boundary conditions used in the micro-scale simulations are presented later, and finally, the generation of computational grids is discussed.

Generation of virtual 3D fibrous structures

In this study, virtual fibrous structures with different fiber orientations, namely unidirectional and isotropic structures, were constructed using GeoDict, a voxel-based commercial code (Math2Market GmbH, GeoDict2012R1, www.geodict.com). In GeoDict, the fiber orientation distribution is controlled by a density function $p(\psi,\varphi)$ defined in polar coordinates as

$$p(\psi,\phi)=\frac{1}{4\pi}\left(\frac{\beta\sin\psi}{(1+(\beta^2-1)\cos^2\psi)^{3/2}}\right), \tag{3}$$

where β is an anisotropy parameter, and its value reflects the fiber orientation of the micro-structure generation, $\psi\in[0,\pi)$ and $\varphi\in[0,2\pi)$ are through-plane and in-plane angles, respectively (Ashari and Vahedi Tafreshi, 2009). The density function (Eq. 3) depends on through-plane angle only, and the fiber orientation is controlled by the value of β. For an isotropic structure, the value of β is equal to one. As the value of β increases, the fibers tend to become parallel to the in-plane. The degree of fiber alignment is described by the second-order fiber orientation tensor Ω (Soltani et al., 2014; Stylianopoulos et al., 2008),

$$\Omega=\frac{1}{l_{tot}}\sum l_i \times\begin{bmatrix}\sin^2\psi\cos^2\phi & \sin^2\psi\sin\phi\cos\phi & \cos\psi\sin\psi\cos\phi\\ \sin^2\psi\sin\phi\cos\phi & \sin^2\psi\sin^2\phi & \cos\psi\sin\psi\sin\phi\\ \cos\psi\sin\psi\cos\phi & \cos\psi\sin\psi\sin\phi & \cos^2\psi\end{bmatrix}, \tag{4}$$

where l_i is the length of the ith fiber, l_{tot} is the total fiber length, and the sum is over all fibers within the domain. The values of diagonal components in the fiber orientation tensor Ω (Eq. 4) are a measure