Microfluidic model of porous media wetting Application to a collagen network

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ABSTRACT

A slightly deformable network may be considered as a particular porous media with a connected void phase. In this paper, a fluid flow through a collagen network is analyzed with applications in the area of restorative dentistry. Infiltration of a fluid into a network already saturated by another fluid is a complex issue especially when the fibers are so tightly distributed (10 to 100 nm between them in a dentinal collagen network) that capillary forces are significant (the bond and capillary number are small).

In a general framework, the equation of the capillary membrane with variable curvature and the associated boundary condition at the contact line between the two fluids and the fiber wall are discussed. The resulting solution is approximated by the finite element method together with a level set technique to track the smeared front.

The expected meniscus shape around a fiber and the capillary membrane advancing front in different networks are obtained. The influence of fiber orientation and interfibrillar space is discussed. This type of simulation is an essential step towards a refined understanding of the macroscopic behavior of a porous medium like the collagen network in order to estimate e. g. the equivalent capillary pressure or relative permeability.

INTRODUCTION

This study stems from a need in restorative dentistry but as it deals with flow under capillary forces, it can be extended to a wide range of areas like soil mechanics. Adhesion to dentin is currently achieved by creating a micromechanical seal between a resin and demineralized dentin. Demineralized dentin is a porous medium made of enlarged tubules and a collagen fiber network uncoverered by demineralization (Figure 1). Practitioners encounter a lack of durability of the seal due to the incomplete and heterogeneous infiltration of the collagen network leading to the degradation of the bond. A better understanding of the infiltration of the collagen scaffold is thus necessary to point out how to improve bonding conditions.

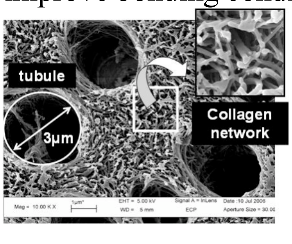

Figure 1: Demineralized dentin two porosities

In this study, we try to model the dentinal collagen tissue geometry and its infiltration. The major challenge is to take into account capillary effects when tracking the front. For this purpose, we chose to use the level set method to track the front since it allows avoiding a special meshing of the front to be tracked. In the same spirit, the fibrils are materialized by cylinders of high viscosity in our model to avoid the same type of meshing difficulties. Thus, the unsaturated porous material is modeled by three fluids, one for the scaffold itself and two for invading and displaced fluid.

In a first section, we review the equilibrium of a capillary membrane in a general framework and point out classical but not necessary assumption and its implementation in a finite element code through a weak formulation of the system of partial differential equations. Then, we present the chosen geometrical model of the dentinal collagen fiber network. Three particular networks are finally considered which allow discussing fibers orientation, interfibrillar space and boundary conditions influence.

THE CAPILLARY MEMBRANE

Capillary membrane local equilibrium

The capillary membrane is the thin interface between two fluids endowed with special mechanical properties. Let σ_f (with f=1,2) be the stress tensor inside each fluid and σ_m the stress tensor of the interface S which is assumed to be like a membrane tensor that is to say with vanishing transverse shear. The superficial equilibrium equation on the interface reads (with the superficial divergence Div_S) and n the unit normal to S pointing from fluid 2 to fluid 1) :

$$Div_S\ \sigma_m + (\sigma_1 - \sigma_2)\ (n) = 0$$

With the classical assumption of an isotropic membrane stress p_m and when the surrounding fluids are at equilibrium, the superficial gradient of the tension p_m vanishes so that this tension is constant over the membrane. Then p_m is given by:

$$p_m = \ ((\sigma_1 - \sigma_2)(n), n) / \ div_S n)$$

from which the classical Laplace's formula can be recovered. It should be remarked here that a constant superficial gradient and isotropic membrane stress are not mandatory as considered in the first equation and have already been considered in the literature.

Variational formulation of the infiltration process

The virtual power principle is written on each fluid (1 and 2) and on the capillary membrane S, with contact line ∂S, and the three equations are added to give (if we choose $w=w_n n$ and $grad_S w_n=0$):

$$\int_{\Omega f} (\rho_f\, a_f, w)\ dV + \int_{\Omega f} Tr(\sigma_f . Dw^T)\ dV - \int_S p_m\ w_n\ div_s n\ dS = \int_{\Omega f} (\rho_f\, g, w)\ dV + \int_{\partial S} p_{m}(\nu, w) dl$$

Let us consider the line integral on the right hand side which is the term that is responsible for the expected meniscus shape (contact angle θ) :

$$\int_{\partial S} p_m(\nu, w)\ dl = \int_{\partial S}\ p_m(\cos\theta\ n - n_w\ )\ dl$$

with n_w the normal vector of the solid wall. The solid wall can be either an exterior boundary or an implicit obstacle like a fiber. Thus, the weak form of the whole system gives:

$$\int_{\Omega f}(\rho_f\, a_f, w)dV+\int_{\Omega f} Tr(\sigma_f . Dw^T)dV-\int_S p_m\, w_n\, div_s n\, dS=\int_{\Omega f}(\rho_f\, g, w)dV+\int_C p_m(\cos\theta n-n_w)dl$$

To sum up, the weak formulation enables to take into account capillarity (through the term $p_m\, div_s n$) and the meniscus shape along the implicit solid walls.

Capillary surface approximation with a level set technique

Actually the interface is an unknown free surface. It is interesting to parameterize it by a function $\varphi(x,t)$ whose a particular level set will represent the interface([1]). Moreover, $\varphi(x,t)=1$ in fluid 1 and $\varphi(x,t)=0$ in fluid 2. The mean position of the interface is given by the isovalue 0.5 of φ. As it is a material surface, the time material derivative $d\varphi/dt$ should vanish. Thus:

$$\partial\varphi/\partial t +(\nabla\varphi, v) = 0$$

The initial conditions are set up to represent a smeared approximation of the initial capillary front (given by φ_0) through a regularized Heaviside function h: $\varphi(x,0)=h(\varphi_0(x),\varepsilon)$. The front is followed via the level set function isovalue and the coupling is done with Navier Stokes equation through the velocity term v. Using $n=\nabla\varphi/|\nabla\varphi|$, the surface integral of the variational formulation is obtained.

Contact line approximation

The contact line ∂S is the intersection between the fiber wall and the capillary surface. If a fixed level set is used to identify the fiber location, actually the same the one used to identify the viscosity of these fibers, the contact line characteristic function is identified as the product of the Heaviside functions of the wall and the capillary surface. The line integral which appears in the variational formulation can then be easily computed over the finite element mesh.

COLLAGEN TISSUE MODELING

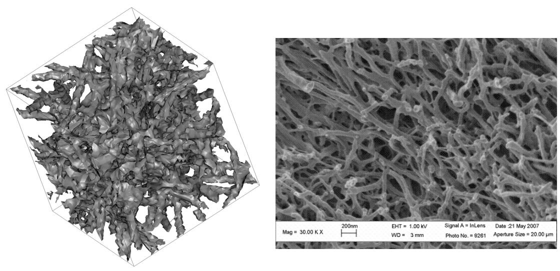

Figure 2 : Modeled and real networks

The collagen tissue deep infiltration is crucial to efficient bonding between a restoration material and the dentinal substrate. The real network made of collagen fibers characteristics and the way we have chosen to model the geometry of the porous medium are now presented.

Implicit modeling of collagen fibers

The collagen fiber network of demineralized dentin has been widely observed especially by transmission electron microscopy, scanning electron microscopy and atomic force microscopy. Fiber diameter is 50-100 nm ([2]) in the hydrated state. Taking into account these morphological observations, the collagen fiber network is approximated by a network made of cylinders which have the same diameter of 80 nm. The real dentinal collagen fiber network porosity has been assessed using mercury porosimetry and is roughly 55% so that the cylinder number can be set to reach this porosity value. However, no quantitative study on fibers orientation has been led although a dependence on localization was observed. As fiber orientation is not known deterministically, we propose to study the orientation influence in the next session.

The cylinders are represented by their characteristic functions so that a sum of regularized heaviside functions is representing the cylinder distribution. Figure 2 illustrates a given realization of fiber random distribution within the control volume compared to the real network on the right hand side.

The model is validated by comparison between the flow around an implicit and an explicit fiber (with an appropriate choice of the viscosity ratio). Flow rates of the same order of magnitude and similar streamlines have been found. Moreover, Darcy's permeability using implicit fibers is in agreement with collagen network permeability values found in other studies ([3]).

INFILTRATION MODELING OF ORIENTED FIBROUS NETWORKS

The resin flow through the network remains to be determined to better understand the infiltration process called "hybridization". By following the transient flow, the influence of fiber orientation and interfibrillar distance are discussed.

Interfibrillar space influence

Vertical fibers network is a simple and convenient type of network to appreciate the effect of fiber distance. So, two networks are built with different distances between five fibers. The distance to the explicit wall (which does not exist in practice) also plays a role.

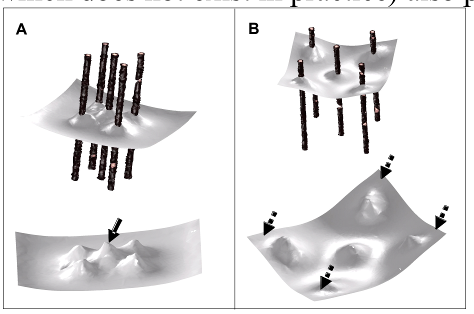

Figure 3: influence of the distance between fibers. A: d=0.8µm. B: d=1.6µm

The figure 3 presents the front shape depending on the distance set between fibers. There seems to exist a threshold distance above which no interaction between fibers occurs. Here when the interfibrillar space d is 0.8 µm (Figure 3A), the meniscus of the central fiber interacts with its neighbors and its meniscus is higher than its neighbors whereas when this distance is increased the interaction decreases (d=1.6 µm, Figure 3B). The solid wall meniscus interacts with fibers when d=1.6 µm so the results are really meaningful in a zone close to the center only. To overcome this numerical artifact, periodic boundary conditions have been used thereafter.

Orientation influence

Three types of network with periodic boundary conditions have been built :

- a network with five vertical fibers randomly set in the control volume (VERT)
- a network with five fibers in plans perpendicular to the front direction (PAR)
- a network with five fibers randomly set in the control volume (no constraint on their position and direction) (RAND)

The results are illustrated by Figure 4. The vertical fibers (VERT) network infiltration is deeply influenced by the distance between fibers. The capillary rise is higher when the fibers are closer (red arrow) so that the conclusion of the previous paragraph is confirmed with periodic boundary conditions that are avoiding meniscus interaction with exterior walls (Figure 4A, black arrow).

In the PAR network the front seems less drawn by capillarity. It is because the capillary membrane and the fiber walls do not always intersect. The dashed arrow shows that fibers are nevertheless attracting the membrane but only when the front is approaching. The black arrow points out an area that stems from the periodic boundary condition.

In the RAND network, the main observation is that the orientation variation of two close fibers may increase drastically the front rise (Figure 4C, red arrow). It can be figure out that a large gradient of orientation may lead to a local acceleration of the infiltration process and consequently to a heterogeneous penetration.

Improving resin penetration

Interfibrillar space is a crucial parameter. As in a capillary tube (where the larger the diameter of the tube, the higher the capillary rise) when the fibers are closer the front rise is higher. But in our case, this space cannot be lowered that much because then the molecules of the resin are too large to creep through it.

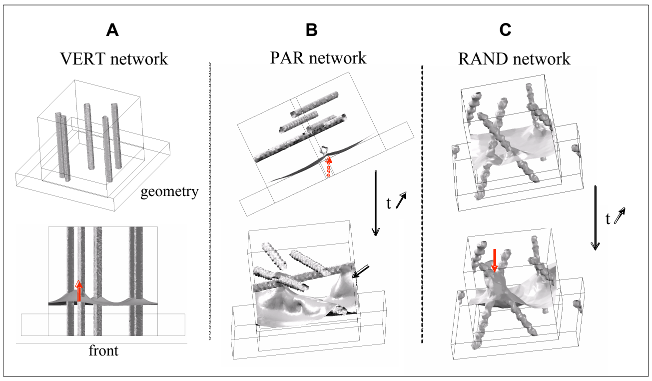

Figure 4: Advancing front through the three different networks

The capillarity effects influence more the flow through a vertical fiber network than through a network with fibers perpendicular to the front direction because the boundary condition term that gives the expected meniscus shape is active only when the front is passing through fibers. This term is always active in VERT networks. So, it is believed that modifying the network into a VERT network can enhance infiltration. Using this idea, Pasquantonio et al. ([4]) is applying an electric field to demineralized dentin and shows that it is improving bonding. One of the possible reasons could be that the field is reorienting the network into a VERT network.

CONCLUSION

In this paper, fiber networks have been infiltrated by a fluid using the level set method and the Navier Stokes equations with capillary terms. The fronts have been computed for different fibers orientations and their final geometries have been discussed qualitatively. This work will be applied to demineralized dentin collagen network infiltration in restorative dentistry to estimate the heterogeneity of resin penetration essential to the adhesion of the seal.

The main observation is that the front is significantly influenced by fiber orientation and interfibrillar space. We may wonder whether those parameters can be changed to improve dentinal infiltration. Contact angle, surface tension coefficient influence and penetration time are also to be investigated before infiltrating the full network. Estimation of macro properties like equivalent capillary pressure and relative permeability can be performed from the simulation described here.

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