The Role of Relative Permeability in Simulation of RTM Process Filling Phase

Z. Dimitrovová† and S. Advani‡

† IDMEC, Instituto Superior Técnico, Lisbon, Portugal
‡ Department of Mechanical Engineering
University of Delaware, Newark, USA

in B.H.V. Topping, Z. Bittnar, (Editors), "Proceedings of the Sixth International Conference on Computational Structures Technology", Civil-Comp Press, Stirlingshire, UK, Paper 132, 2002. doi:10.4203/ccp.75.132

Abstract

In this paper new aspects are introduced into the numerical simulation of the mould filling phase of the Resin Transfer Moulding process of composites manufacturing. Necessity of transient analyses on three levels is justified and importance of consideration of the theory of wetting (unsaturated) flows through dual porous media, implying inclusion of relative permeability and homogenized capillary pressure at the macrolevel, is stressed. First methodologies for relative permeability and homogenized capillary pressure determination, applicable to macroscopically unidirectional pressure driven viscous flows through porous media are described. They are based on a detailed study of the resin progression performed by a Free Boundary Program developed by the authors. Some numerical aspects of the Free Boundary Program are also discussed.

Keywords: resin transfer moulding, dual porous media, free boundary flows, relative permeability, capillary pressure, surface tension, Stokes flow, Darcy's law

1 Introduction

RTM (Resin Transfer Moulding) is a widely used process to manufacture advanced composites with continuous fibre reinforcements. The process starts by an arrangement of fibre bed in a mould cavity, which is followed by injection of a thermoset resin into the closed and clamped mould. In the next stage, by increasing the mould temperature, the liquid resin cures and solidifies and after that the part is demolded in the final stage. From manufacturing viewpoint, one would like to fill the spaces between the fibres before it starts curing, because the viscosity of the resin increases rapidly with degree of cure and high viscosity might aggravate or even stop resin flow leaving voids and dry spots, which are detrimental for the mechanical resistance of the final part [1]. The relatively new RTM process still have many phenomena, which are known to occur in RTM applications, but they are

still not well understood neither theoretically nor experimentally. Semi-analytical models for absolute permeability establishment, the role of capillarity, methodologies for relative permeability and macroscopic capillary pressure determination are subjects of current research.

2 Macrolevel analysis

2.1 Standard approach

The RTM filling phase can be numerically modelled as an independent problem, with the main objectives to determine progression of the resin front and evolution of the pressure distribution, to predict dry spots and voids formation and to optimise injection gates and air vents locations that will accomplish successful impregnation. Filing time is one of the main factors determining the overall cost of the fabricated part, because during filling both, the resin in a deposit as well as the mould, must be kept at exactly determined and constant temperature, ensuring to keep the resin temperature during filling as much as possible constant and at the value guarantying the lowest viscosity. Due to the fully controlled external heating, energy equation may not be included in the governing equations of the resin motion and resin viscosity can be assumed as constant.

Due to the thousands of fibres and very low ratio of their characteristic cross-sectional size to the size of the mould, numerical simulation of the filling phase, accounting for all the fibre geometry details, is a formidable task and not necessary. Homogenisation techniques are often used in order to reformulate the original problem in terms of the microlevel (local) and the macrolevel (global, effective) analyses. Flow of commonly used resins can be viewed as low Reynolds number flow of an incompressible Newtonian liquid, therefore the macrolevel problem is described by Darcy's law for incompressible flow through porous media, [2-6]. The resistance of the fibre preforms to the flow is lumped into a (absolute) permeability tensor, which is required as one of the input data. One could either obtain it experimentally or predict it by analytical or numerical methods by averaged velocity of Stokes flow in fully saturated basic cell to represent the repetitive fibre architecture.

At the macrolevel, resin motion is commonly assumed as quasi-steady state flow. In standard approaches resin front is represented as sharp discontinuity. In each time step pressure field is solved for (under steady-state conditions) and then explicit schemes with sufficiently small time increase are adopted to model the movement of the resin into the fibre preform according to frontal velocities, [7-8]. Moreover, resin flow is promoted only on middle surface of the part and in-plane components of absolute permeabilities for various fibre layers are replaced using the rule of mixtures. Transversal permeability components do not enter the analysis at all. Such approach can only detect macroscopic filling problems, like large dry spots.

Infiltration of the low viscosity thermoset resin (usually only 50 to 5000 times more viscous than water) into empty spaces of a stationary fibre preform is driven

by the hydrodynamic pressure gradient originated by the higher inlet pressure, however, in certain regions this gradient can be so low, that the wicking gradient will exceed it and the driving mechanism of the flow will change. This is plausible when preforms built from fibre tows (containing few thousands of fibre strands) are used, Figure 1. In such preforms, the spacing between the tows can be an order of magnitude higher than the spacing of pores inside the tows, forming in this way not single but dual porous media. These preforms allow reaching much higher fibre volume fractions and consequently ensure better mechanical properties of final composites and are therefore overly preferred over the traditional reinforcements.

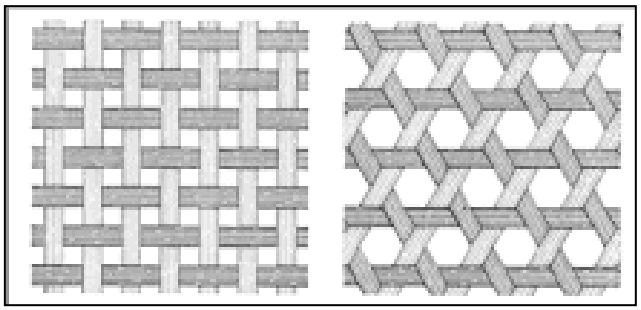

Figure 1: Fibre layers composed by fibre tows

Roughly speaking, high hydrodynamic pressure gradient promotes the resin in the inter-tow spaces, while when wicking gradient is dominant, resin proceeds more quickly through intra-tow spaces. These phenomena imply non-uniform resin front at the microlevel, which translates into transition (partially saturated) region along the macroscopic resin front, which is often clearly visible and which the standard approaches cannot capture.

Similar phenomenon is observed even with traditional reinforcements across the thickness. In Figure 2 velocities and pressure distribution across the thickness is shown for specimen made of two layers with different in-plane permeabilities. Two options for transition region definition are marked. Either it can be considered as a region where saturation is strictly between 0 and 1, or it can include also the region where the flow does not still have the steady-state profile.

2.2 Modifications of the macroscopic governing equations

In order to describe the transition region, new variable saturation, s, varying from 0 to 1 and defined as the ratio of the filled pore space to the total pore space in a basic cell, must be introduced into macroscopic analysis. Saturation introduction implies modification of the continuity equation (mass conservation) to:

$$\phi \frac{\partial s}{\partial t} = -\nabla \cdot \mathbf{v}^{D} , \qquad (1)$$

where $\mathbf{v}^D$ is the phase averaged velocity called as Darcy's velocity, ϕ is the porosity of the preform and t stands for time. Since $\mathbf{v}^D = s\phi\mathbf{v}^f$, where $\mathbf{v}^f$ is the intrinsic phase

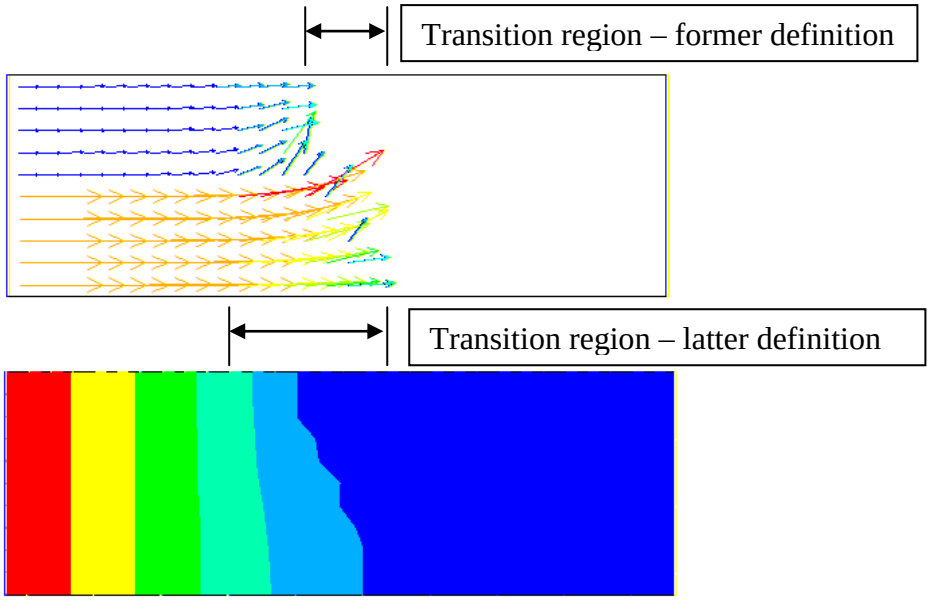

Figure 2: Two different definitions for the transition region

averaged velocity, spatial derivatives of the saturation are required making exact implementation of this equation difficult.

However, Equation (1) is often used together with Darcy's law for steady-state flows:

$$\mathbf{v}^{D} = -\frac{\mathbf{K}}{\mu} \cdot \nabla P \,, \tag{2}$$

where **K**, P and μ are the absolute permeability tensor, macroscopic pressure and resin viscosity, respectively. The macroscopic counterpart, P, of the local pressure is the intrinsic phase averaged value. This approach, used e.g. in [9], only means different approach to the resin front tracking and no capillarity is accounted for. Other attempts use single porous medium model, standard approach and tow filling is included as an additional sink term [10-11].

Few attentions have been paid so far to the theory of flow through dual porous media, even if such studies could discover some injection problems and consequently prevent voids or dry spots formation. Modifications of Darcy's law have to include two more new functions. First of all macroscopic pressure should be divided into its hydrodynamic part, P, and capillary part, $P_c(s)$, [12], and absolute permeability tensor has to be replaced by effective permeability tensor, $\mathbf{K}^{ef}$, in which each component is multiplication of relative permeability, $k_{ij}(s)$, and absolute permeability component K_{ij}. This is suggested in [13, 14], however without accounting for anisotropy as here:

$$\mathbf{v}^{D}(s) = -\frac{\mathbf{K}^{ef}}{\mu} \cdot \nabla\left(P(s) - P_c(s)\right), \tag{3}$$

Besides the macro and the microscale, a mesoscale, with characteristic dimension of the fibre tow diameter, is required to implement. Absolute permeability calculation in dual porous media can be accomplished by mesolevel analysis, in the same way as in single porous media. New functions of saturation, relative permeability k and macroscopic capillary pressure P_c, must enter the analysis as known functions. Equation (3) is used e.g. in [15], but k(s) and $P_c(s)$ are introduced in form of functions determined experimentally in other fields of applications, in soils mechanics. Fibre preforms have, however, completely different architecture, they are highly anisotropic and the resin flow is viscous. Therefore these curves should not be used in composites manufacturing. Another form of Equation (3) can be derived by averaging of the local momentum equation for periodic media [4], but in this case also the resin front must have a periodic pattern, which is very restrictive.

Unlike absolute permeability, no simple procedure is available to determine relative permeability or macroscopic capillary pressure at least numerically. Macroscopic capillary pressure should correspond to the average of its microscopic counterparts, $P_c(s) = 2\gamma\langle H\rangle(s)$, where H is the mean curvature and γ stands for surface tension, which is also not easy to include generally. Any methodology of k and P_c determination has to be based on transient micro and mesolevel analysis. This was our motivation to create the Free Boundary Program (FBD).

3 The Free Boundary Program

FBD enables to study the filling process at the micro or mesolevel. It uses the general-purpose finite element code Ansys to calculate a base analysis and it is written in the Ansys Parametric Design Language and Fortran. Current stage is still only two-dimensional, but it has full capability of capturing the dynamics of fluid motion, it includes surface tension influence, contact angle formation and substitution of fibre tows by single porous medium, when applicable. During its development it was already presented in [16-18]. Other approach to model transient mesolevel analysis is in [19], it involves Lattice Boltzman method as a numerical approach and Brinkman equation in intra-tow spaces as one of the governing equations, but the methodology there does not offer all capillarity features, which can be included in FBP.

Also in FBD, filling is simplified as quasi-steady state process and explicit time integration is used in the free boundary condition for the front progression. Moving mesh scheme is adopted, in order to model correctly the front curvature. Besides FBD being a base of semi-analytical methodologies for relative permeability and macroscopic capillary pressure, it permits one to specify resin properties and fibre architecture for an acceptable filling.

3.1. Microlevel analysis

At microlevel, resin flow obeys Stokes law and at the free boundary the following conditions must be fulfilled:

$$\boldsymbol{\sigma}^{v}\cdot\mathbf{n}=\mathbf{0}, \tag{4}$$

$$p=-p_c=-2\gamma H \tag{5}$$

$$\partial f/\partial t+\mathbf{v}\cdot\nabla f=0, \tag{6}$$

where $\boldsymbol{\sigma}^{v}$ is the local viscous stress, $\mathbf{n}$ is the unit normal vector, p and p_c stand for local pressure and capillary pressure, respectively, function $f(\mathbf{x}(t),t)=0$ describes the moving front and $\mathbf{v}$ stands for local velocity. Besides the fact that (4-6) must be fulfilled, no-slip condition must be imposed at the fibre boundary and contact angle must be formed at the resin front-fibre contact point.

In FBP, first, at each frontal nodal point at a current time step, t_k, the velocity is extracted and a local coordinate system (called 1-lcs as shown in Figure 3) is created close to it, where the flow front is locally approximated by a smooth curve, which includes two adjacent nodal points.

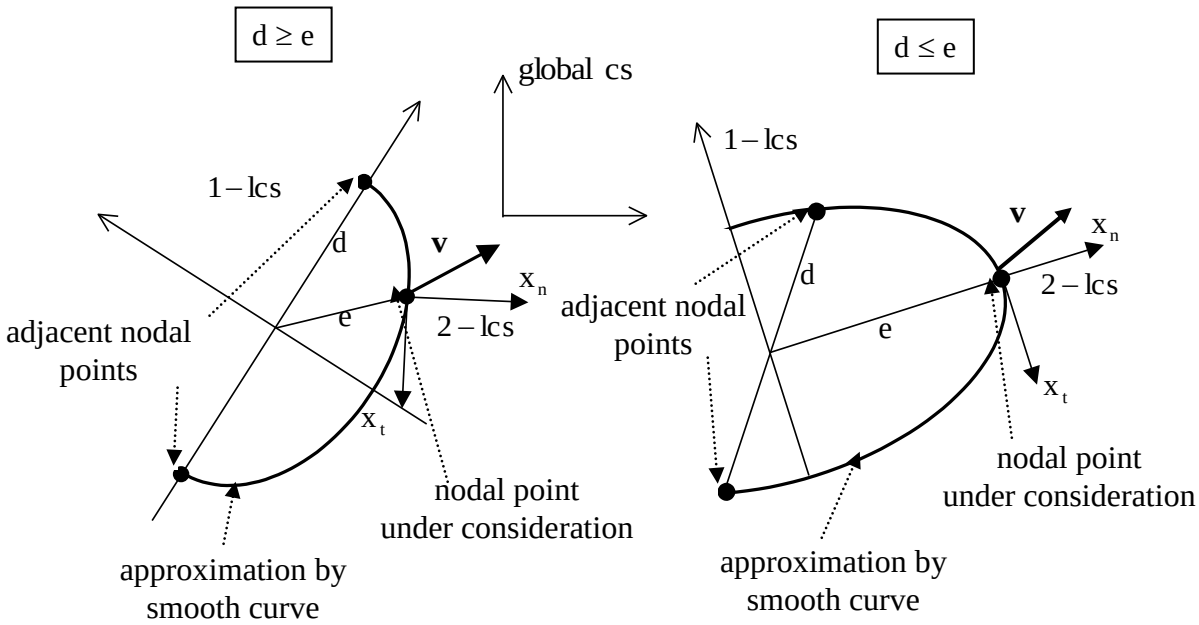

Figure 3: Two kinds of approximation of the flow front by a smooth curve

This permits to define uniquely the outer normal vector to the flow front. Two different ways to approximate are explained in Figure 3. An elliptic approximation in lieu of a circular or a parabolic one is preferable. Next, a second local coordinate system is created at each nodal point with axes along the tangential, x_t, and the outer normal vector, x_n (2-lcs). If the flow front is described locally by $x_n=g(x_t)$ with respect to this coordinate system, then Equation (6) can be written as:

$$v_n - v_t\frac{\partial g}{\partial x_t} = v_n = \frac{\partial g}{\partial t}\,. \tag{7}$$

In the explicit approach, the new nodal position is determined in the normal direction as:

$$x_{n,t_{k+1}} = x_{n,t_k} + (t_{k+1} - t_k) v_{n,t_k} . \tag{8}$$

The smooth curve approximation as described above is repeated, now for the new front, and the surface curvature is determined in order to calculate the capillary pressure, which is then applied according to (5) as piece-wise linear at each frontal element edge. Finally other boundary conditions are applied and Stokes problem is solved for in the new domain.

There is no need of special treatment of the singularity point at the resin front-fibre contact. In progression without the contact angle formation resin progression along the fibre surface is ensured by the neighbouring point motion (Figure 4a). If contact angle is formed, in FBP additional circular surface is created (Figure 4b) in order to adjust exactly the given angle. This radius as well as the contact angle are accurately measurable characteristics, depending on the properties of the three phases: resin-fibre-air, and must enter the analysis as known parameters.

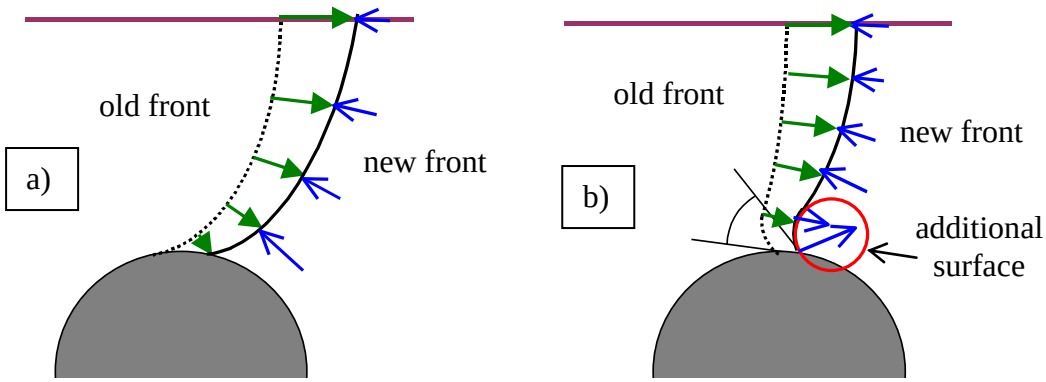

Figure 4: Front progression without (a) and with formation of the contact angle

Adjustment of the resin front to the contact angle is very important, in order to ensure the correct total value of the capillary pressure applied at the front surface, because capillary pressure acting on an arbitrary part of a free surface satisfies equivalent load conditions with the surface tension applied in its cuts. Therefore, the total capillary force, which should be applied at the resin front, can always be determined analytically if the resin front-fibre contact point locations and the parameters γ and θ are known. In numerical simulations dependence on the additional radius was found as insignificant, however, differences were discovered between the total predicted and applied capillary force. Since in the older version adjustment of the contact angle was done only approximately, now exact location of the contact point is calculated in Maple, procedure of which is called directly by FBP.

Viscous flow across cylindrical fibres is strongly influenced by capillary number N_c, defined as $v\mu/\gamma$, where v is a characteristic velocity of the studied problem. FBP

simulation results confirm this fact by results of front progression in Figure 5. It is seen that as capillary number decreases, capillarity becomes stronger and the resin front approaches the constant curvature surface, as derived analytically in [20].

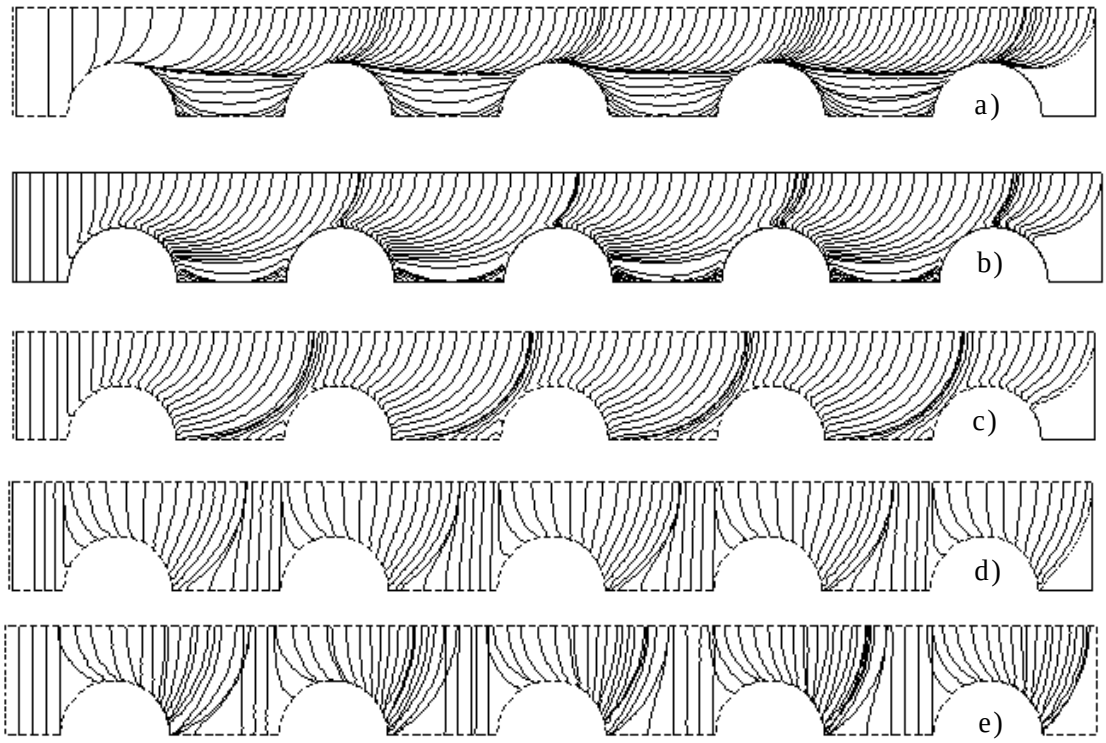

Figure 5: Flow across cylindrical fibres with circular cross section for capillary numbers ∞, 2.76, 0.276, 0.0276, 0.00276, respectively, results obtained by FBD

At microlevel, analogy with incompressible elasticity can be exploited, which has the advantage of linear base analysis. In example of flow across cylindrical fibres with elliptical cross section and N_c=0.05, it is verified, that the front progression for both kinds of base analysis, incompressible elasticity and fluid dynamics (Flotran module of ANSYS), is exactly the same, Figure 6.

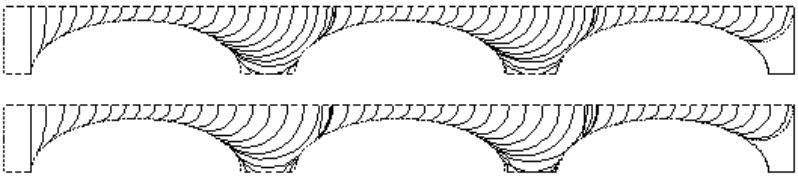

Figure 6: Front progression for flow across cylindrical fibres with elliptical cross section, base analysis as incompressible elasticity (above) and fluid dynamics (below).

However, maximum velocity is slightly different (Figure 7) and mass conservation, i.e. preserving of flow rate at the outlet is better fulfilled in structural than in fluid dynamics analysis.

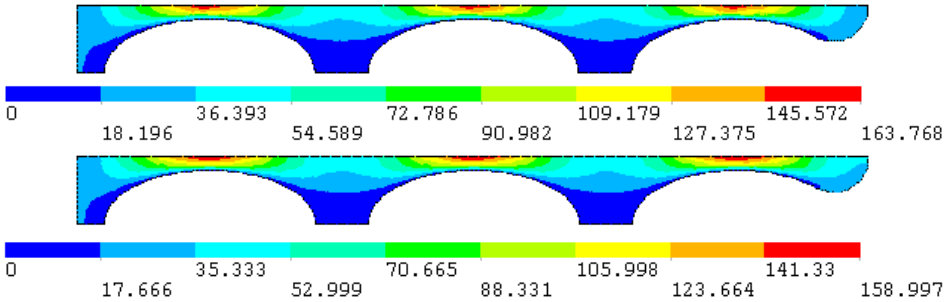

Figure 7: Horizontal component velocity distribution in flow across cylindrical fibres, base analysis as incompressible elasticity (above) and fluid dynamics (below).

3.2. Mesolevel analysis

At mesolevel, Stokes flow is promoted in inter-tow spaces and Darcy's flow in intra-tow spaces. Free boundary condition in intra-tow spaces has different form, as it has already to include averaged fields:

$$\partial f / \partial t + \left(\mathbf{v}^{t} \cdot \nabla f\right) / \phi_{t} = 0 , \qquad (9)$$

ϕ_t is intra-tow porosity and $\mathbf{v}^t$ represents the phase averaged velocity on basic cells related to fibre strands in intra-tow space. In Flotran analysis porous medium is introduced by distributed resistance by introduction of intra-tow permeability. Capillary pressure acting in intra-tow spaces does not depend on front curvature, but its value can be sufficiently estimated from number of contact points at fibrils surface, contact angle, surface tension and averaged position of the front along the fibrils surface, as can be justified at the microlevel.

Verification of newly included fluid dynamics (Ansys Flotran module) base analysis at mesolevel is done firstly on saturated flow. In Figure 8 velocity pattern and pressure distribution are shown for fiber tow with 7000 fibrils and intra-tow porosity 0.4. The expected patterns and rapid decrease in velocity values inside the tow is confirmed.

Flow front progression and the corresponding currently filled total space with different colour designating the tow space are shown in Figure 9-10, for capillary number equal to 0.025 and 0.05, respectively, and for the same characteristics of fiber tows as in previous example, leading the estimate of capillary pressure $1.9 \cdot 10^{-3}$ MPa. Dimensions of mesolevel basic cell are 2×1 mm and the tow cross section is elliptical, in conformity with the reality. It is seen that the tow filling is significantly delayed. Care must be taken of the tow surface, because here the velocity jumps rapidly to very low value, almost as if discontinuity were presented. Therefore,

inclusion of nodal point at the tow surface into the front progression might affect the flow pattern and oscillation of the bulk front could be detected. FBD includes special treatment to eliminate these points from the free boundary condition.

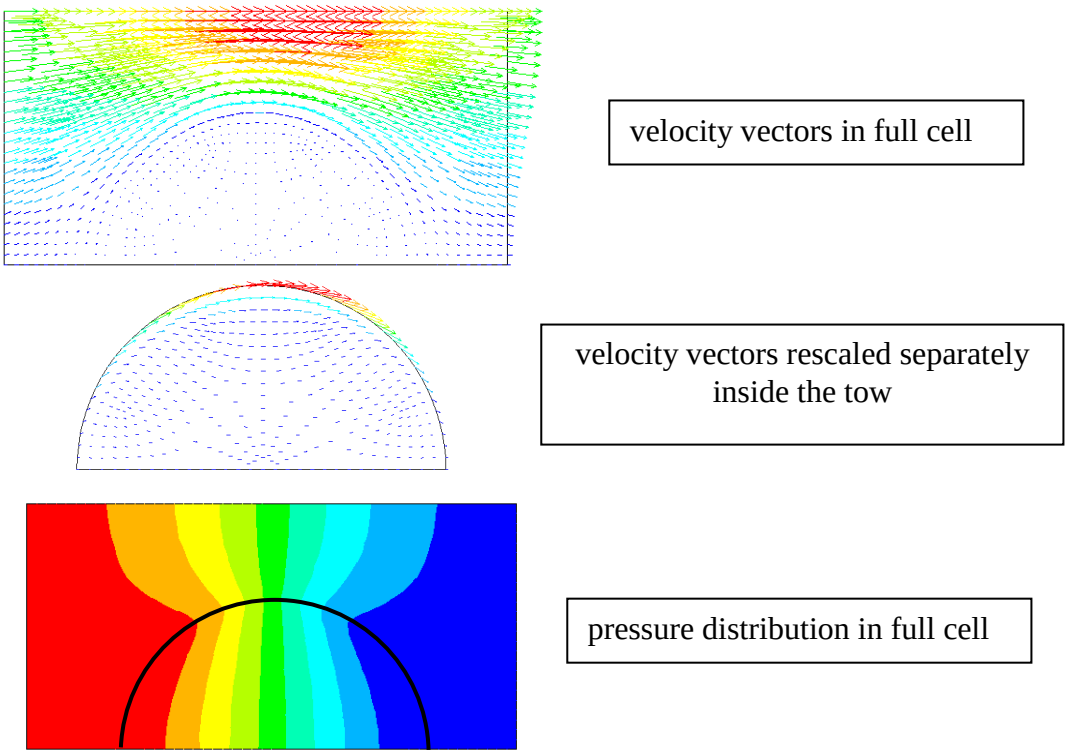

Figure 8: Velocity and pressure distribution in a saturated basic cell

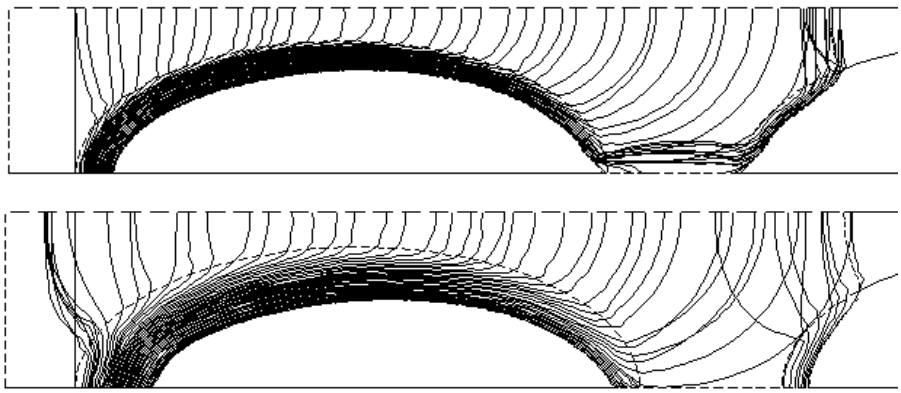

Figure 9: Flow front progression for N_c=0.05 and 0.025

Although the front progression looks very reasonable in studied examples, mass conservation was not exactly satisfied. This is not FBD problem, but Flotran "incorrectness" of the non-linear algorithm giving the fluid dynamics results not truly steady-state. This phenomenon must be further investigated.

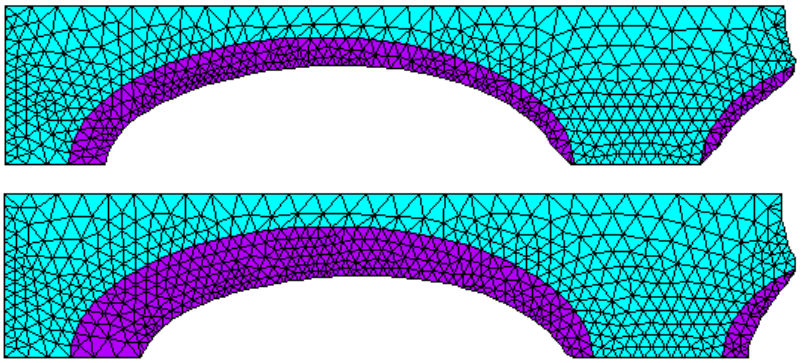

Figure 10: Currently filled total space with different colour designating the tow space

4 Methodology for relative permeability determination

Practically no methodologies applicable to composite manufacturing for the relative permeability and/or macroscopic capillary pressure are available, first attempt derived by authors of this paper is described in [17-18]. It is applicable to macroscopically unidirectional filling and requires possibility of introduction of uniform basic cell. Uniform basic cell can be defined as a cell in which during the resin infiltration the saturation increases from 0 to 1, while the previous cell is fully saturated and the next cell is empty. If the flow permits uniform cell introduction, then under assumption, that after the uniform basic cell is saturated, local velocity and pressure distribution will have immediately a periodic pattern, relative permeability can be derived in the form:

$$k_{xx} = \frac{G \cdot \varepsilon_s}{G + \frac{2A_{P_c,\varepsilon_s}}{\varepsilon_s}\left(1-(2\varepsilon_s-1)^m\right)} \tag{10}$$

where A_{P_c,ε_s} is the mean value of the capillary pressure, ε_s is dimensionless spatial designation of the flow front position and G<0 is the periodic pressure gradient. Equation (10) is obtained by averaging of the necessary characteristics over all possible uniform cells. All inputs of Equation (10) can be obtained without the necessity of studying the particular progression. Besides the geometrical parameters, k_{xx} strongly depends on the capillary number N_c. Results for flow across cylindrical fibres with circular cross section and relative radius of 0.25 for different capillary numbers are presented in Figure 11.

Unfortunately uniform basic cell cannot be introduced at the mesolevel in cases reported in Figures 9-10 and therefore Equation (10) is not applicable to these flows.

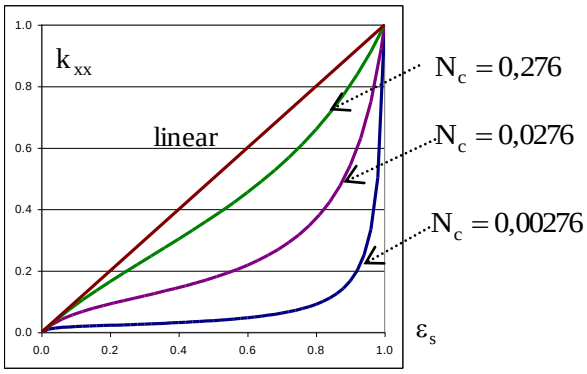

Figure 11: Dependence of relative permeability on capillary number

5 Conclusion

FBD is found as a useful toll in studying viscous flow progression at micro and mesolevel, permitting to account for all capillarity features. It permits one to specify resin properties and fibre architecture for an acceptable filling. Further work must be done in order to include three-dimensional analysis. It is also necessary experimental verification of the obtained results.

Described methodology for relative permeability is very simple to use and it permits to conclude that relative permeability depends on both, surface tension as well as viscous force, which might be supported by the fact, that resin front progression at the microlevel strongly depends on capillary number. Unfortunately it is not applicable to common cases of mesolevel filling. Further extension of the methodology is necessary in order to account for other filling situations.

References

[1] S.G. Advani, M.V. Bruschke, R.S. Parnas, "*Resin Ttransfer Molding*", in Flow and Rheology in Polymeric Composites Manufacturing, Ed. S.G. Advani, Elsevier Publishers, Amsterdam, 465-516, 1994.

[2] A.E. Scheidegger, "*The Physics of Flow through Porous Media"*, The Macmillan Company, New York, 1957.

[3] J. Bear, "*Dynamics of Fluids in Porous Media"*, Dover Publications, New York, 1972.

[4] M. Kaviany, "*Principles of Heat Transfer in Porous Media"*, Springer-Verlag, New York, 1995.

[5] E. Sanchez-Palencia, "*Non-Homogeneous Media and Vibration Theory"*, Lecture Notes in Physics, 127, Springer-Verlag, 129-157, 1980.

[6] H.I. Ene, D. Polisevski, "*Thermal Flow in Porous Media"*, D. Reidel Publishing Company, 1987.

[7] M.V. Bruschke, S.G. Advani, *"A Numerical Approach to Model Non-Isothermal, Viscous Flow with Free Surfaces Through Fibrous Media"*, International Journal of Numerical Methods in Fluids, 19, 575-603, 1994.
[8] B. Liu, S. Bickerton, S.G. Advani, *"Modeling and Simulation of Resin Transfer Molding (RTM) - Gate Control, Venting, and Dry Spot Prediction",* Composites A, 27A, 135-141, 1996.
[9] A. Shojaei, S.R. Ghaffarian, S.M.H. Karimian, *"Numerical Simulation of Three-Dimensional Mold Filling Process in Resin Transfer Molding Using Quasi-Steady State and Partial Saturation Formulations",* Composites Science and Technology, 62, 861-879, 2002.
[10] W. Chang, N. Kikuchi, *"Analysis of Composite Molding Using the Homogenization Method",* Computational Fluid Dynamics, 7, 49-77, 1996.
[11] K.M. Pillai, *"Flow Modeling in Dual Scale Porous Media"*, Ph. D. dissertation, The University of Delaware, Delaware, USA, 1997.
[12] N. Patel, V. Rohatgi, L.J. Lee, "*Micro Scale Flow Behavior and Void Formation Mechanism During Impregnation through a Unidirectional Stitched Fiberglass Mat",* Polymer Engineering and Science, 35, 837-851, 1995.
[13] M. Muskat, *"The Flow of Homogeneous Fluids through Porous Media"*, New York, NY:McGraw Hill, 1937.
[14] R.E. Collins, *"Flow of Fluids through Porous Media"*, Reinhold Publishing Corporation, 1961.
[15] K. Han, "*Analysis of Dry Spot Formation and Changes in Liquid Composite Molding"*, Ph.D. theses, The Ohio State University, 1994.
[16] Z. Dimitrovová, L. Faria, *"Finite element modeling of the RTM process"*, Proceedings of the 5th International Conference on Flow Processes in Composite Materials, Plymouth, UK, July 1999, 125-135.
[17] Z. Dimitrovová, S.G. Advani, *"A Semi-Analytical Approach to the Relative Permeability Determination in LCM Processes"*, Proceedings of the 8th International Conference on Composites Engineering, Tenerife, Canary Islands, Spain, August 2001, 191-192.
[18] Z. Dimitrovová, S.G. Advani, *"Analysis and Characterization of Relative Permeability and Capillary Pressure for Free Surface Flow of a Viscous Fluid across an Array of Aligned Cylindrical Fibers",* Journal of Colloid and Interface Science, 245, 325-337, 2002.
[19] M.A.A. Spaid, F.R. Phelan, *"Modelling void formation dynamics in fibrous porous media with the Lattice Boltzman method",* Composites A, 29A, 749-755, 1998.
[20] P. Concus, R. Finn, *"Capillary Surfaces in Microgravity",* in Low-Gravity Fluid Dynamics and Transport Phenomena, Ed. J.N. Koster and R.L. Sani, Progress in Astronautics and Aeronautics, 130, American Institute of Aeronautics and Astronautics, 183-205, 1990.