Void Transport in Resin Transfer Moulding

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Abstract

Void formation in Resin Transfer Moulding (RTM) has, during the latest years, been a subject of considerable attention. Less attention has been given to the transport of formed voids. The voids may move with the resin but they may also be trapped in the reinforcement. Here, void transport is discussed through industry scale experiments, model experiments with capillary tubes, injections carried out under microscope and theoretical computations. The results are applied on real situations occurring in RTM and it is found that than a converging injection strategy is preferable when minimising the void contents. It is furthermore found that low permeable areas are areas where voids can be trapped.

1. INTRODUCTION

Resin Transfer Moulding is a well known processing method for fibre reinforced polymer composites. Although the properties of manufactured composites are generally quite good, they are often inferior to what is potentially possible since several flaws may be introduced during the manufacturing process [1], [2]. One type of defect is voids, i.e. gas bubbles occupying part of the matrix area. It has been found that the shape and the location of the voids can vary considerably. In RTM the voids are usually divided into cylindrical voids, located within the fibre bundles, and more spherical voids located between the fibre bundles [3].

Several ways to minimise the void content has been presented [3], [4], [5] where the two most effective are vacuum assistance during mould filling and an applied pressure during cure. With both these methods it is possible to manufacture composites without voids. However to be able to give advice on processing parameters in order to minimise the void contents for specific cases a deeper understanding of void formation and transport is required.

This report will start with a discussion of the preform geometry followed by a review of work carried out on void formation. Then void transport will be considered through some model experiments and the results obtained will be applied on real situations occurring in RTM.

2. PREFORM GEOMETRY

Preform geometry plays an important role for the formation and transport of the voids. The fibre mats utilised in RTM consist of bundles of fibres, which are stitched, wowen, braided or held together by some sort of binder. Each bundle can contain hundreds or even thousands of fibres and may span over the whole composite, or be chopped with a length of a few centimetres. In Fig. 1 a few examples of fibre mat structures are shown. As seen, the structures of the mats are quite complicated, as the different channels formed between the bundles may have both different directions and cross-sectional areas. In the preforms of RTM, the fibre mats are quite frequently laid in various directions, or different types of fibre mats are utilised. In complicated mould geometry the reinforcement may be formed over corners, be stretched, sheared and compressed to different thicknesses. This complicates, even more, the geometry of the preform as seen on the fibre bundle scale.

Within a fibre bundle, randomly distributed cylindrical fibres are observed, oriented mainly in the same direction; cf. Fig. 2. Between the fibres, channels are formed with different size due to the non-even distribution of the fibres. The cross-section of the channels will vary slightly *along* the fibres since they may be twisted around each other. By observing the geometry in a plane *perpendicular* to the fibres, one may envisage channels for which the cross-section varies considerably; cf. Fig. 2.

The two scales described above, the fibre bundle scale and the fibre scale, are generally of little interest to the manufacturer of RTM parts. Rather, properties of the preform on the mould scale, such as the fibre fraction and the permeability of the preform are of interest. Geometrical variations of the preform can also be identified on this scale. This applies, for instance, when the number of fibre mats changes or when another type of reinforcement is used in some part of the mould.

3. VOID FORMATION

It is generally accepted that voids are formed at the resin flow front due to irregularies of the preform on different scales: i) the mould scale ii) the fibre bundle scale and iii) the fibre scale.

On the mould scale, variations of the permeability throughout the mould may lead to formation of large air inclusions, so called dry spots. The resin flow front moves faster in high permeable areas and air may be enclosed in the low permeable areas as shown in Fig. 3. A similar result can be obtained if the resin viscosity varies throughout the mould. For more complicated geometry of the preform than the one in Fig. 3, dry spots may form if a bad choice of injection strategy is made. To be able to predict and avoid dry spots, both simple analytical expressions for mould filling have been derived [6], and mould filling simulation programs have been developed [7], [8]. The continuous filling of the mould when a dry spot is formed, may result in small air bubbles being released from the dry spot which then will follow the resin through the inter-bundle channels in the reinforcement; cf. Fig. 3.

On the fibre bundle scale (the intermediate scale) the quite complicated geometry may result in a considerable void formation. One inter-bundle channel can be wider than another which may end in air being entrapped in the smaller channel. This mechanism does not seem to have been studied in connection with composite manufacturing. However, the situation is similar in oil recovery. By scrutinising the pore-doublet model, see Fig 4, Rose and Whitherspoon [9] concluded that the displacing fluid (the resin) may move faster in the larger channels enclosing the displaced fluid (the air) in the smaller channel.

Another cause for void formation is the large difference in flow resistance in the space between the fibre bundles and within the bundles. Theoretical and numerical models of flow through fibre reinforcements have shown that voids can form at the resin flow front [10], [11]. Parnas and Phelan [10], for instance, considered the flow perpendicular to cylindrical fibre bundles and found that the flow into the bundles is considerably slower than the flow around the bundles. Hence, air is trapped in the bundles. Experimental studies have also shown that air may be enclosed both within the bundles and between them, depending on where the resin flow front moves the fastest. At high speeds of the flow front the resin preferably moves in the channels between the bundles, while at low injection speeds the capillary pressure within the bundles dominates the flow [12]. In the high speed case, air is trapped within the bundles, while in the second case the air is enclosed between the bundles. An optimum speed of the flow front should therefore exist, when the resin moves at equal rate in the high and low permeable areas [12], [13].

Within the fibre tows, the resin flow front may also move faster in some areas than in others. The consequence of this is that cylindrical shaped voids are formed within the bundles. Except for geometrical aspects, uneven surface treatment of the fibres may influence the formation of voids on the fibre scale.

Besides being created at the flow front, there is a possibility that voids move into the reinforcement. Firstly, there might be voids in the resin, before it is injected into the mould. Secondly, voids may form from air, which is drawn into the mould during injection or cure. This is the case if the pressure is lower in the mould than outside and if the mould is not properly sealed. The latter cause was put forward as one possible explanation, if poor results are obtained with vacuum assisted RTM, [4].

During the injection, but particularly at the curing stage of the process it is a risk that voids are generated by nucleation, if the local gas or moisture contents in the resin is too high and the pressure inside the mould is too low. This subject has been thoroughly investigated for the auto-clave process. Kardos et. al. [14] defined, for instance, safe areas (no nucleation) as to processing parameters for prepregs stored at different relative humidity. Other causes for voids to appear in the manufactured composite arise when they are formed by: i) cavitation due to resin shrinkage or ii) chemical reactions within the resin, or between the resin and the surface treatment of the reinforcement.

4. VOID TRANSPORT

In addition to the formation of the voids the transport of them has a large influence on the final void contents and distribution in the composite. During the processing, enclosed gas (or volatile components in the resin) may move as voids or as dissolved molecules. One indication of such movements was reported in [3]. Laminates with different lengths were manufactured under identical processing conditions and by letting the resin flow from one side of the mould to the other (parallel flow). Hence, the shorter injections can be seen as snapshots of the longer injections. In Fig. 5 the void contents of two of the laminates (0.2 and 0.55 m long, respectively) are shown. It is observed that for both injections the voids are concentrated at the outlet side of the mould. A comparison between the void distributions for the two laminates shows clearly that the voids move in the flow direction of the resin. The local speed of the flow and the geometry of the reinforcement are both important parameters for the transport of the voids. A different scenario for the voids located within the bundles and those located between the bundles is therefore expected. General rules for both types of voids are:

i) The voids are influenced by the pressure, as given by the perfect gas law. In [4] the volume of a void at position ξ was, for isothermal conditions, expressed as:

$$V_\xi = \frac{p_{out}}{p_\xi} V_0 \tag{1}$$

where p_{out} is the outlet pressure and V_o is the initial volume of the void. This relationship was used in order to explain the improved quality of the composite, when the vacuum level was increased, [4]. A *critical volume* when the voids get mobile was defined and when V_ξ reaches this volume the void will escape the entrapment. Eq. (1) does not account for the capillary pressure which in most cases may be neglected, but for small bubbles the capillary pressure, should be added to both pressures in (1).

ii) The *critical volume*, as described in [4], must be correlated to the processing conditions in some way. For instance, voids will escape the entrapment in the fibre reinforcement if the pressure gradient is high enough. The following expression was found in [15] to model bubbles moving through a constricted capillary tube:

$$\Delta p = \frac{2\gamma_{lv}\cos\theta}{R_{con}}\left(1 - \frac{R_{con}}{R_v}\right) \tag{2}$$

where Δp is the pressure over the bubble, θ the contact angle between the gas-liquid-solid interface and where R_v and R_{con} are the void and constriction radii, respectively. Low surface tension, γ_{lv}, of the liquid and uniform channels in the reinforcement will make it easier for the bubbles to move with the resin. The smaller the scale studied is, the higher pressure gradient will be required to force a bubble through a constriction. Hence, for the same ratio between the void radius and the constriction radius voids are more likely to be trapped within fibre bundles than between them, [15]. In this paper it was also experimentally derived that the detailed geometry of the constriction does not affect the required pressure.

iii) In (1) it is neglected that the voids will increase or decrease in size due to molecule transport over the gas-resin interface. Effects of this were monitored from injections carried out under microscope, [16]. Without any increase in pressure it was observed that entrapped voids decreased in size. A quantification of the decrease of void size inside fibre bundles carried out in [17] yields that the length of the voids changes as:

$$l = l_0 e^{-\frac{2GD_r H\left(1-p_0/\left(p_\xi + p_c\right)\right)}{R_v^2 \ln\left[\left(R_v + R_s\right)/R_v\right]\rho_{atm}} p_{atm} t} \qquad (3)$$

where l_0 is the initial length of the void and where the indices *atm*, *o* and ξ for the pressure denote atmospheric pressure, degassing pressure and pressure at position ξ, respectively. In the exponent are included also a geometrical constant G, the diffusion coefficient D_r of the specific gas in the resin, a constant H which multiplied by the pressure gives the saturated concentration gas in the resin, the density of the gas at atmospheric conditions ρ, the void radius R_v and the thickness of a stationary region R_S. Eq. (3) is derived from studies on cylindrical voids and change in volume changes only their length. Still the equation may also be used to show the trends for the more spherical inter-bundle voids.

In the inter-bundle channels the motion and the diffusion of the bubbles are set by the flow within these channels. Since the resin flow rate here is often much higher than within the bundles, [10, 17], the spherical bubbles can move fast towards the resin flow front or change in volume faster, due to diffusion, relative to the cylindrical bubbles. The bubble can, however, be trapped at constrictions in the resin channels or be pushed away into stagnant flow areas. One way for the bubbles to escape their entrapment is to decrease in size and thereby become more mobile. Consider a void formed and trapped near the inlet of the mould. The pressure will increase as the flow front moves into the mould and the void will consequently decrease in size both by the perfect gas law and by an increased diffusion rate; cf. (1) and (3). When its radius, R_v, becomes small enough the void will move through the constriction; cf. (2). As the void moves towards the resin flow front, the pressure around the void will decrease with a corresponding increase of the void volume (1). But also the tendency for the gas to dissolve into the resin will be reduced; cf. (3). The void may then be trapped again, shrink, escape and move to a new constriction, etc. A concentration of the voids towards the resin flow front is therefore to be expected. This was also measured for all cases presented in [3, 4]; cf. Fig 5.

Within the fibre bundles the flow rate is considerably lower than outside the bundles, [10, 17]. and the speed of the voids is correspondingly lower. Although it has been observed that some of the voids in the bundles move into the inter-bundle channels, it is common that the voids stay trapped within the bundles, [16]. From (2) it can be derived that high pressure gradients are required to force the cylindrical voids out of the fibre bundles. From measurements of void contents in cured laminates it was also found that the relative contribution of the cylindrical voids increased from 38% near the outlet side of the laminate to 100% 0.25 m inside the laminate, [3]. Observations made on injections carried out under microscope qualitatively give the same results, [16]. The voids that stay trapped can only disappear by being compressed as predicted by the perfect gas law, and by molecular diffusion. The alteration in volume of the cylindrically shaped bubbles only changes their length and they must become very small, before they can escape their entrapment. The higher the pressure is, the faster the voids will become sufficiently small to totally dissolve into the resin.

5. DISCUSSION OF THE RESULTS

The studies presented in the previous Section point to conditions that are preferable for transport of the voids. High pressure gradients for transport of the voids, or high pressure for dissolution of the voids, are such examples. When aiming for low void contents it is desirable to optimise the process with respect to these parameters. A manufacturer, for instance, has to choose injection strategy which for nearly all geometries can be chosen from either of edge injection (line injection), point injection or peripheral injection, (cf. Fig. 6), or from a combination of them. The pressure distribution for the three cases are computed from [18]:

$$\nabla^2 p = 0 \qquad (4)$$

with the boundary conditions given in Fig. 6 and with the assumption that the preform has uniform permeability, that the thickness of the mould is constant, that the resin has constant viscosity and that the flow follows Darcy's law. For the same materials and processing parameters the different strategies will give different pressures; see Fig. 7.

As discussed previously, the volume of the voids is affected by the pressure in two ways. i) directly by the perfect gas law (1) and ii) indirectly by the dependence from the pressure on the diffusion rate (3). An increase in pressure will decrease the volume of the void, which may reduce the required pressure gradient to force a void through a constriction (2). The dissolution of the void may also lead to a bubble being released and it might, too, result in the bubble being totally dissolved into the resin. Hence, the higher the pressure throughout the mould is, the less voids are expected. As shown in Fig. 7 (and neglecting the end point of the curves where the pressure is equal for all three strategies) the pressure is highest for peripheral injection throughout the mould, while it is lowest for point injection.

From Fig. 7 also the difference in pressure gradient can be studied. Near the flow front the pressure gradient is highest for peripheral injection (notice that the flow is in the negative *r*-direction for peripheral injection). Somewhere in the middle of the mould, edge injection has the highest pressure gradient while at the inlet the pressure gradient is highest for point injection. Consider a bubble formed and arrested in some constriction near the flow front. If the chosen strategy is peripheral, the bubble will directly feel a high pressure gradient and the possibility for the bubble to move towards the flow front is much larger than for the other two strategies. A bubble located near the inlet is more likely to move for the point injection case. However, since the pressure gradient decreases towards the resin flow front, the bubble may be trapped in another constriction in the reinforcement before it reaches the resin flow front. Regarding the advection of the bubbles, peripheral injection seems to be most preferable, although there is a risk that bubbles are arrested near the inlet side. It is thus concluded that, a converging injection strategy seem to be a better alternative than a diverging injection strategy when optimising the process for low void contents.

According to the experimental observations in [3] the voids will be located near the resin flow front. This is the case for injections where the permeability is uniform in the flow direction. However, when the permeability varies in this direction, the pressure gradient will also vary and bubbles may be trapped at locations with low pressure gradients. Consider, for instance, the case in Fig. 8a. Following Bear [18] the ratio between the pressure gradient in area A and area B is approximately given by:

$$\frac{G_B}{G_A} = \frac{2K_1}{K_1 + K_2} \qquad (5)$$

Let K_1 be a typical permeability for a fibre mat in RTM, ($5 \cdot 10^{-11}$ m^2) and let K_2 represent the permeability for an open two dimensional channel with a height of 1 mm ($3 \cdot 10^{-7}$ m^2). For this simple case the ratio in (5) is $3 \cdot 10^{-4}$. Thus, bubbles that have moved into area B, where the pressure gradient is very low, may easily be trapped in constrictions; cf. (2). Fig. 8b shows two examples where this can be a reality in real RTM mouldings: i) when the number of layers are changed and ii) when the reinforcement is partly compressed, for instance, in corners where an open space can be left in the outer corner.

7. References

1. Holmberg, J.A., Lundström, T.S., Gebart, B.R., Strömbeck, L.A., "Process Induced Defects in Resin Transfer Moulding", *SICOMP Technical Report 94-005*, Swedish Institute of Composites, Box 271, S-94126 Piteå, Sweden, (1994)

2. Summerscales, J., "Manufacturing Defects in Fibre - Reinforced Plastics Composites" Submitted for publication

3. Lundström, T.S., Gebart B.R., "Influence from Different Process Parameters on Void Formation in RTM", *Polymer Composites*, 15, pp. 25-33 (1993)

4. Lundström, T.S., Gebart B.R., Lundemo, C.Y., "Void Formation in RTM", *Journal of Reinforced Plastics and Composite*, **12**, pp.1340-1349 (1993)

5. Hayward, J.S., Harris, B., "The Effect of Vacuum Assistance in Resin Transfer Moulding", *Composites Manufacturing*, **1**, pp. 161-166 (1990)

6. Gebart, B.R., Lundemo, C.Y., Gudmundson, P., "An evaluation of alternative injection strategies in RTM", *Proc.of the 47th Annual Conference of the Society of Plastics Institute*, Cincinatti, USA (1992)

7. Fracchia, C.A., Castro, J. and Tucker, III, C.L., "A Finite Element/Control Volume Simulation of Resin Transfer Mold Filling", *Proc. of the American Society for Composites Fourth Technical Conference*, pp. 157-166, Technomic, Lancaster, PA (1989)

8. Koorevaar, A., "Simulation of the Resin Transfer Moulding (RTM) Process", *Proc. of the Techtextil - Symposium*, Lecture No. 338 (1995)

9. Rose, W., Witherspoon, P.A., "Trapping Oil in a Pore Doublet", *Producers Mothly*, Dec (1956)

10. Parnas, R.S., Phelan Jr., F.R., "The Effect of Heterogeneous Porous Media on Mold Filling in Resin Transfer Molding", *Sampe Quarterly*, Jan, pp. 53-60 (1991)

11. Chan, A.W., Morgan, R.J., "Modelling Preform Impregnation and Void Formation in Resin Transfer Molding of Unidirectional Composites", *Sampe Quarterly*, April, pp.48-52 (1992)

12. Patel, N., Lee, L.J., "Effects of Fibre Mat Architecture on void Formation and Removal in Liquid Composite Moulding", *Polymer Composites*, **16**, pp. 386-399 (1995)

13. Krupp, J., "Verfahrenstechnische Untersuchungen zur Minimierung von Oberflächenfehlern bei großflächigen RTM-Bauteilen", *Institut Für Kunststoffverarbeitung*, D-9225 (1992)

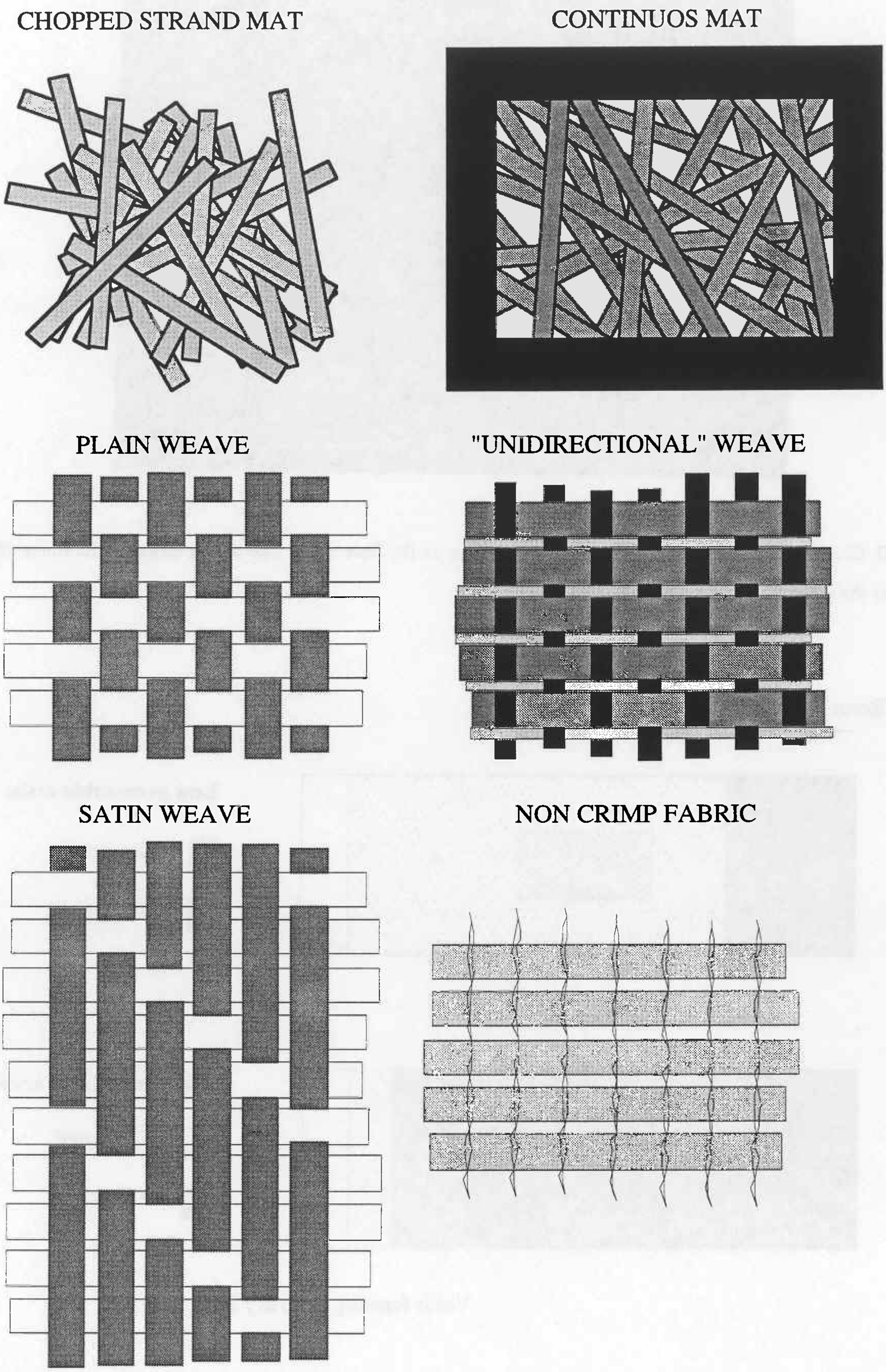
14. Kardos, J.L., Dave, R., Dudukovic, M.P., "Voids in Composites", *Proc. ASME Manufacturing International '88*, pp. 41-48, Atlanta, Georgia (1988)

15. Lundström, T.S., "Bubble Transport Through Constricted Capillary Tubes with Application to Resin Transfer Moulding" *Accepted for publication in Polymer Composites*

16. Lundström, T.S., "Void Formation and Transport in the RTM Process", *Proceedings from the 39th International Sampe Symposium April 11-14*, Anaheim CA (1994)

17. Lundström, T.S., "Effect of Flow on Void Collapse in Resin Transfer Moulding" *Submitted for publication to composites*

18. Bear, J., *Dynamics of Fluids in Porous Media*, Dover Publications Inc., New York (1988)



CHOPPED STRAND MAT

CONTINUOS MAT

PLAIN WEAVE

"UNIDIRECTIONAL" WEAVE

SATIN WEAVE

NON CRIMP FABRIC

Fig. 1. Some patterns for fibre mats. The filled areas are fibre bundles. The two upper figures illustrates two types of random mats. The next three sketches are different types of weaves and the last figure is an example of a stitched fabric.

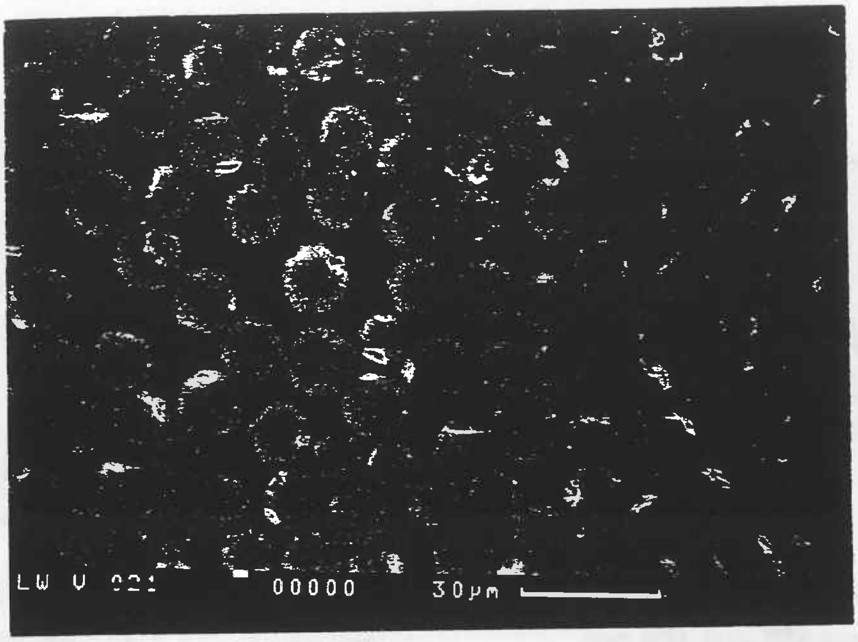


Fig. 2. Cross-section which shows the fibre distribution on the fibre scale. The lighter areas are the fibres and the darker the matrix.

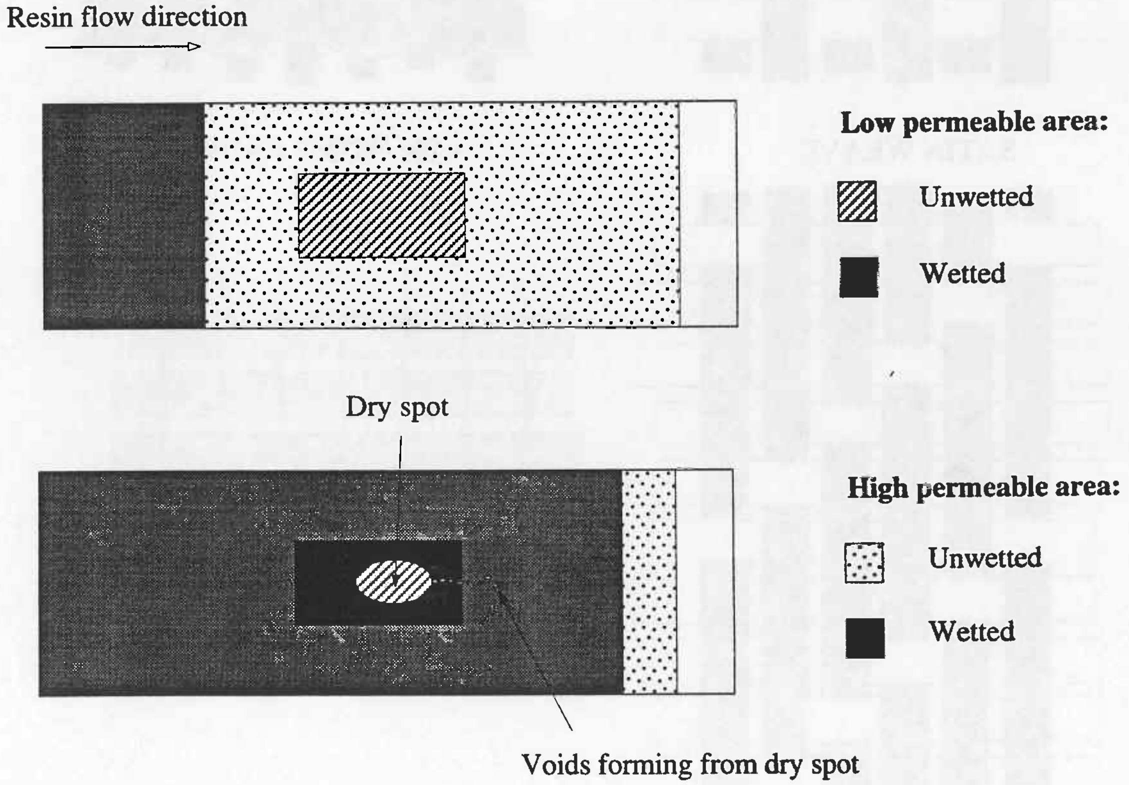


Fig. 3. Example of formation of dry spot. Voids may form from the dry spot and can be transported through the reinforcement.

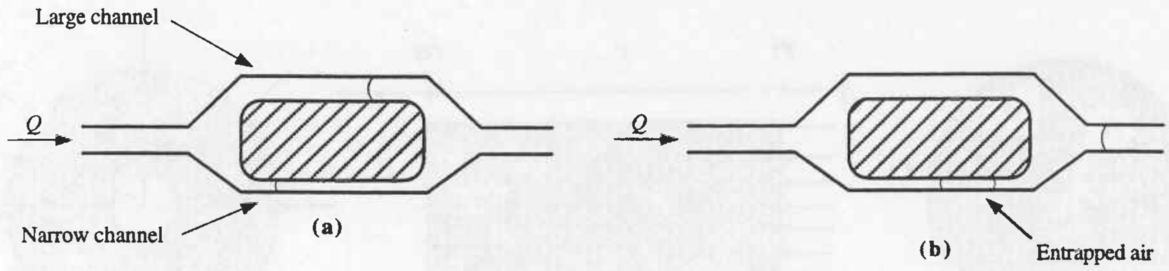


Fig. 4. The pore doublet model. The liquid moves faster in the larger channel and consequently air is entrapped in the smaller channel.

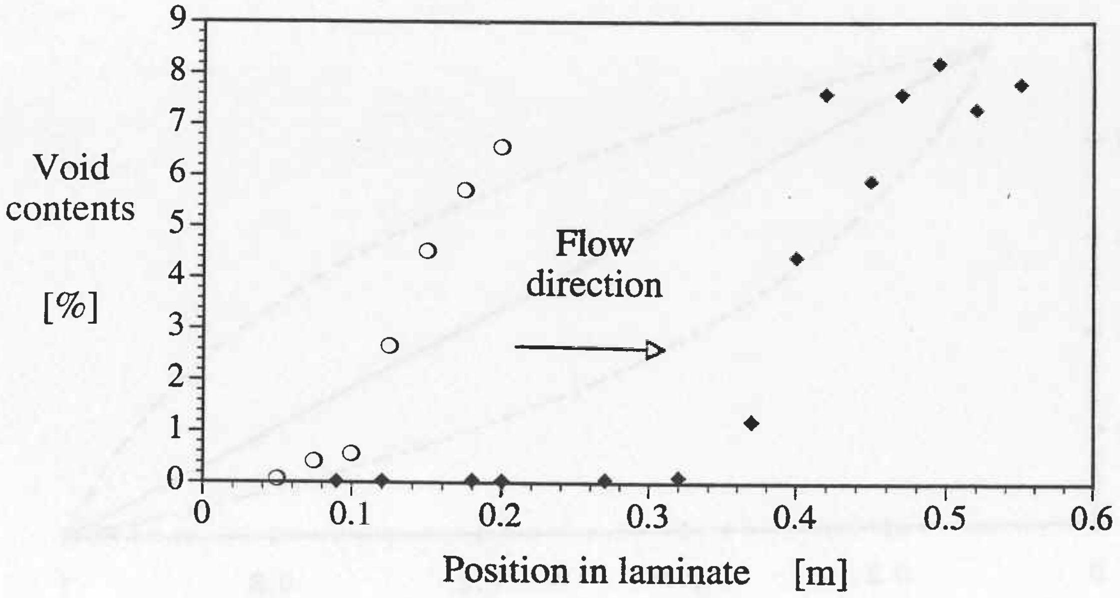


Fig. 5. Void contents measured from two laminates with different lengths. The symbols ○ and ◆ denote laminates with lengths 0.2 m and 0.55 m, respectively.

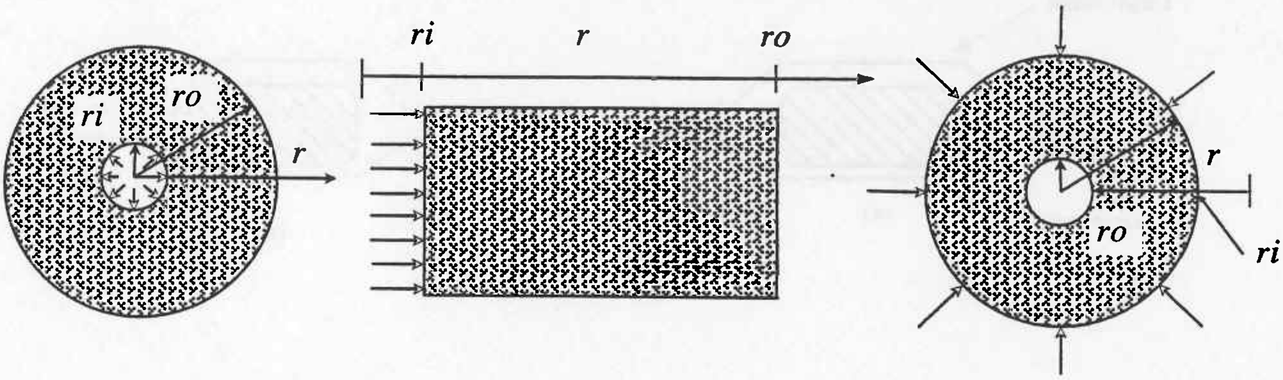


Fig. 6. The three main injection strategies from left to right: Point injection, edge injection and peripheral injection. The open arrows idicates where the pressire p_i is applied. Notice that for peripheral injection the flow is in the negative *r*-direction.

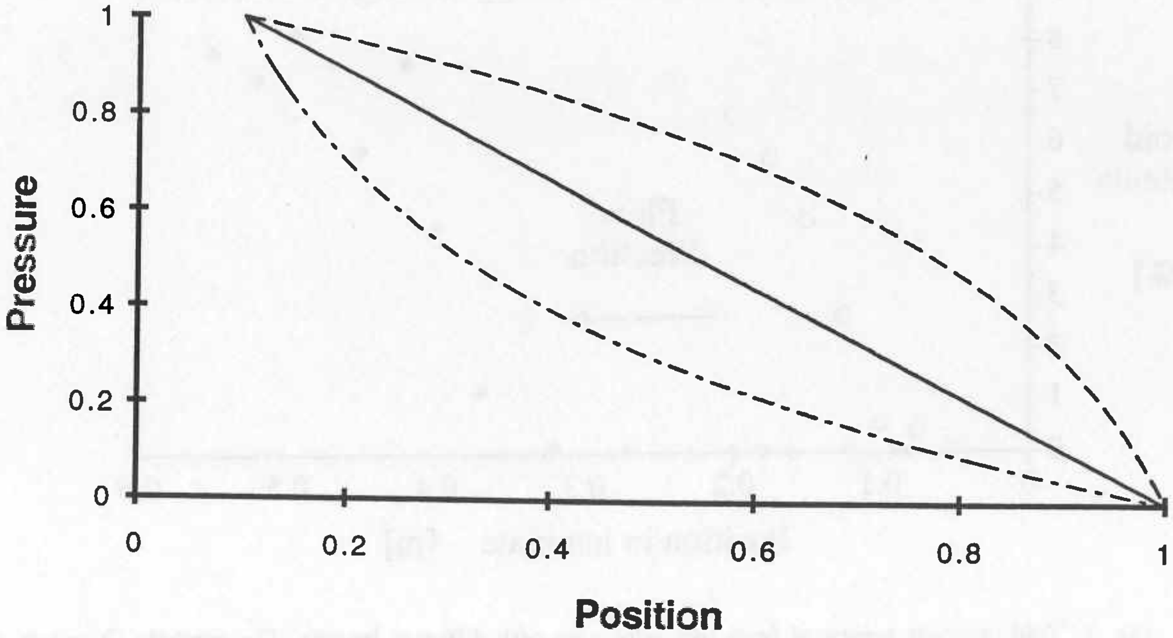


Fig. 7. Dimensionless pressure as function of dimensionless *r*-position for the three injection strategies. The dot-dashed line; point injection, the solid; edge injection and the dotted peripheral injection.

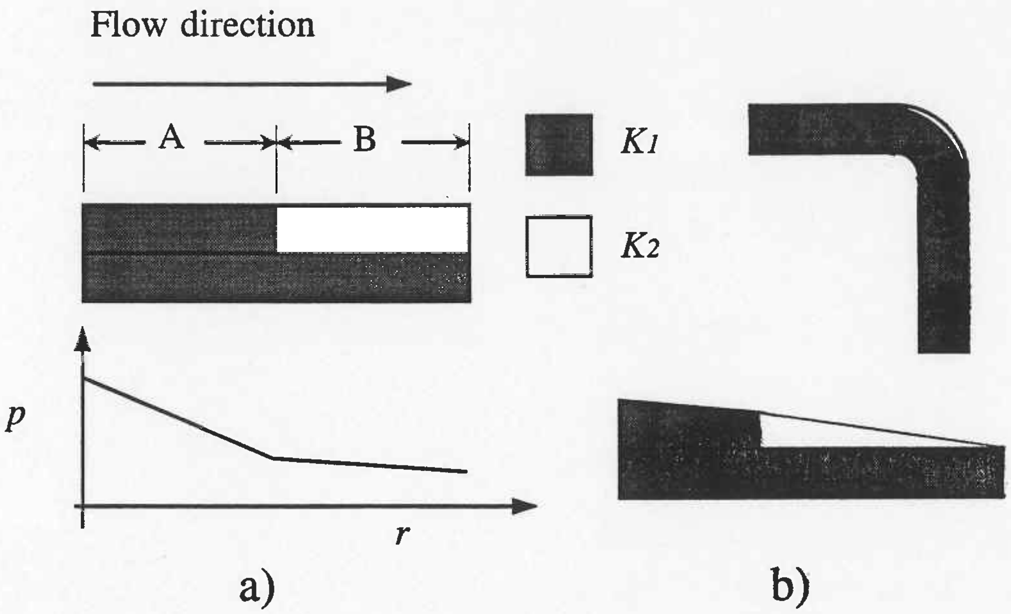


Fig. 8. Examples of geometry with variable permeability in the flow direction.