A Generalised Version of Darcy's Law, and its Use in the Interpretation of Shear Effects in Thermoplastic Melt Pultrusion.

A.J. Longmuir, N. Witten, H.W. Chandler, and A.G. Gibson.

ABSTRACT

Darcy's law for the flow of fluids in porous media is frequently used to describe the flow of resins in beds of fibre reinforcement. An extension is proposed to permit the modelling of the longer range viscous forces which result from externally applied shear flows encountered in thermoplastic melt pultrusion. This allows the decay of velocity perturbations due to die and pin surface effects to be modelled. In profiles of thick section, decay of the velocity perturbation takes place exponentially over a characteristic distance equal to the square root of the Darcy permeability. The model allows the equivalent resin film thickness at the die and pin surfaces to be calculated, and in addition forms the basis of a new method for measuring permeability under processing conditions.

INTRODUCTION

Recently, the manufacture of thermoplastic composites has become possible which has led to some changes in the engineering approach to processing and fabrication. A number of methods have been used to achieve impregnation with thermoplastics but one of the most attractive and convenient is considered to be direct melt impregnation. This method involves the drawing of fibre tows over one or more cylindrical pins in the presence of liquid resin as depicted in Figure 1. This technology is well established for the processing of thermosetting matrix composites where a resin impregnation bath is used prior to forming by filament winding or pultrusion. However, there have been few in-depth studies of the mechanisms by which impregnation takes place. This may be because relatively void-free impregnation can be achieved without the need for significant process optimisation due to the low viscosities of thermoset resins, which are in the region of 0.2 to 2 Pa.s. as compared with thermoplastic melts which are between 10 and 100 Pa.s. Due to this greater difficulty of fibre impregnation with thermoplastic resins there is a need for an improved understanding of the process.

The aim of impregnation processes is to achieve good fibre wetting with controlled resin volume fraction at the fastest possible throughput without undue pressure build-up in the fibres. With pin impregnation, as discussed by Bijsterbosch et al[1] for instance, it is generally necessary to have several pins to achieve adequate resin dispersion. There is a progressive build-up of fibre tension from pin to pin, and a further increase is caused by the wipe-off die. The result of these effects is that line-speed is currently limited by the tension build-up in the various stages of the process. There is, therefore, considerable incentive for process optimisation to achieve effective impregnation with the lowest possible level of tension build-up.

This need for optimisation has led to considerable efforts to obtain realistic models for fabrication operations such as pultrusion. The flow of resin within pores of a 'bed' of reinforcement, in nearly all processes, is seen as the key factor. Darcy's Law has been successfully used to describe the flow of fluids in a porous media. However there are certain types of surface boundary conditions which are common in composites processing which are not easily introduced. In thermoplastic melt pultrusion the resin and the fibre bed are moving firstly along the surface of a stationary pin, and secondly along the surface of a stationary die. If the section is thick enough, the resin at some distance from the pin or die moves at uniform speed. However, the resin in contact with the surface of the die is stationary and this produces a velocity perturbation that extends for some distance into the section. The two main factors which influence the velocity profile into the fibre bed are the pressure gradient and the bulk movement of the fibre bed.

Because Darcy's Law describes only flows which are directly pressure induced it is not possible to introduce boundary conditions which will effectively model the local shear induced boundary interaction present in pultrusion. It is the aim of this paper to propose a modification to Darcy's law to enable the shear effects occurring in melt pultrusion to be modelled.

THEORY

A more comprehensive description of this modification of Darcy's law is given in Reference 2 with respect to modelling mould interface effects. The behaviour of a Newtonian fluid in a regime where inertia forces can be neglected is described by the creeping flow version of the Navier-Stokes equations, expressed in vector form by

$$\nabla p = f + \eta \nabla^2 v \qquad (1)$$

where *f* represents any body force vector within the fluid, and in many creeping flow cases, can be neglected. In the usual form of Darcy's Law a fluid flows in the interstices of a rigid porous medium and the longer range viscous forces are often small in comparison to the local resistance to flow offered by the passages within the material. We can regard this local resistance as a body force *f* so that Darcy's law can be written

$$-\eta \boldsymbol{S}^{-1} \cdot v = f \qquad (2)$$

where ***S*** is the permeability tensor. In cases where longer range viscous effects are significant, Equation 1 then becomes

$$\nabla p = \eta \left(\nabla^2 v - \boldsymbol{S}^{-1} \cdot v \right) \qquad (3)$$

When longer range viscous forces from bulk flow are negligible then

$$\nabla p = \eta \left(\boldsymbol{S}^{-1} \cdot v \right)$$

This is the familiar form of Darcy's Law which, relates the flow velocity vector to the local pressure gradient and the permeability of the porous medium to flow.

Equation 3 is similar to that obtained by Brinkman,1947[3] who developed a model to describe the viscous flow through a dense swarm of spherical particles embedded in a porous mass. Making the assumptions that v_x does not vary in either the y direction or the z direction, and that $\mathrm{d}p/\mathrm{d}x$ does not vary in the y direction, the general solution is found to be

$$v_x = A\exp\left(y/\sqrt{S_{xx}}\right) + B.\exp\left(-y/\sqrt{S_{xx}}\right) - \frac{S_{xx}\,\mathrm{d}\,p}{\eta\,\mathrm{d}\,x} \tag{4}$$

where A and B are arbitrary constants.

Application to melt pultrusion
For both the problems to be described here it may be assumed that the pressure-driven velocity term in Equation (4) is small enough to be neglected. With this assumption, and bearing in mind that v_x is the fluid velocity relative to the stationary surface. This gives

$$v_x = -U \quad \text{at} \quad y = 0$$

and

$$v_x = 0 \quad \text{at} \quad y = \infty$$

where U is the velocity of the fibres relative to the stationary surface. This gives:

$$v_x = -U\exp\left(-y/\sqrt{S_{xx}}\right) \tag{5}$$

The justification of the second boundary condition, which leads to this simple form of velocity decay is given in Reference 2. It can be seen that the surface velocity perturbation decays into the bulk of the bed over a characteristic distance $\sqrt{S_{xx}}$

Equivalent film thickness
A convenient way of describing the viscous drag at the solid surface is to imagine that it is equivalent to that which would be produced by a surface film of neat resin of constant thickness. The thickness of this equivalent film has often been assumed to be comparable with the radius or with the inter-face spacing. Using Equation (5) the value of the equivalent film thickness can be found more accurately in terms of the permeability S_{xx}, since the shear rate in this equivalent film is U/t_{eff}. Therefore by differentiating Equation (5) the shear rate in the resin phase and hence the shear rate at the die or pin interface is given by

$$\left(\frac{\mathrm{d}\,v_x}{\mathrm{d}\,y}\right)_{(y=0)} = \frac{U}{\sqrt{S_{xx}}}$$

The effective film thickness is therefore given simply by

$$t_{\mathrm{eff}} = \sqrt{S_{xx}} \tag{6}$$

EXPERIMENTAL

To investigate the shear effects for both the wipe-off die and the pins in thermoplastic melt pultrusion some measurements were carried out on the impregnation of E-glass tows and polypropylene melt. Three E-glass fibre tows of 2400 Tex were pulled through a pin impregnation bath at 220°C in the presence of a low viscosity polypropylene melt. The pin radius was 10mm and the side restraints limited the tow spreading to a width of 20mm. The die bed was instrumented to allow the pull force transmitted to the pins to be recorded. By varying the geometry of the resin bath the wrap length around the second pin could be varied enabling the wrap length, L_P, to be changed. Four processing speeds of 1, 3, 5 and 10m/min were used. The results of this experiment are shown in Figure 3, in the form of plots of pull force against the wetted surface area of the pins for each processing speed. The use of different wrap lengths allowed the pressure build-up due to the presence of a wipe-off die after the pins to be corrected for. The gradient then gave the shear stress at the surface of the pin at each processing speed. A plot of the resultant shear stress versus line speed is given in Figure 4.

Similarly in the case of the wipe-off dies the pull force was recorded as three tows of E-glass tows of 2400 Tex were pulled through a pin impregnation bath and then through a wipe-off die of constant cross-section. The pull force due to the die was recorded. Wipe-off dies of different lengths were used so that the pressure build-up at the die entry could be corrected for. Dies with three different cross-sections of 2.5, 3, and 3.5 mm diameter were investigated to ascertain the effect of fibre volume fraction on the shear behaviour. Again four different processing speeds of 1, 3, 5 and 10m/min were used. The results of this experiment, at a processing speed of 1m/min, are shown in Figure 5. It is in the form of plots of pull force against the wetted perimeter of the die for the different cross-sections used. Using the same method as above, the shear stresses at the surface of the die at each line speed were calculated from the gradient of the plots. This procedure was carried out on the results from all three sets of dies of different cross-section. The computed results are shown in Figure 6; they are in the form of plots of pull-force transmitted to the wipe off dies, versus line speed for each of the different die cross sections.

The equivalent film thickness was determined from the relationship

$$\gamma = \frac{U}{t_{eff}}$$

where γ is the shear rate, which is determined from the flow curve of the polymer as the shear stress is known, and U is the line speed. The calculated results are given in Table 1 for the pins, and Table 2 for the wipe-off dies. To enable the results of this investigation to be directly compared to the permeability results of other investigations a dimensionless value obtained by dividing the effective film thickness by the fibre radius was calculated. This was then plotted against fibre volume fraction. Similarly the permeability results from other key investigations were divided by the fibre radius and plotted on the same graph. This is shown in Figure 7.

RESULTS

As to be expected the pull force was found (Figure 3) to increase with an increase in the wetted wrap area of the pins and an increase in line speed. The shear stress can be seen in Figure 4 to increase with line speed. There is a fourfold increase in shear stress at the surface of the pin with an increase in processing speed from 1m/min to 10m/min. As with the pins, the pull force due to the wipe-off die is found to increase with an increase in the wetted perimeter of the die and line speed. A more dramatic increase in pull force is seen with an increase in the fibre volume fraction (Figure 5). The same trends are shown in shear behaviour at the surface of the die with processing speed and fibre volume fraction (Figure 6). The comparison of the equivalent film thickness results with the previously reported permeability results is interesting (Figure 7). The results for the wipe-off die at high volume fractions and the pin investigations are found to correlate quite strongly with those of the key paper by Williams et al[4] and show an even greater correlation with the more recent investigation of Rudd and Bulmer[9]. The permeabilities, and hence the equivalent film thicknesses, were found to decrease with an increase in fibre volume fraction.

DISCUSSION

The main benefit of this generalisation to the Darcy's equation is that it makes it possible to determine the extent to which surface shear velocity perturbations penetrate into the bulk material. In many situations the characteristic distance $\sqrt{S_{xx}}$ may be small enough either to be regarded as negligible or to be modelled as an equivalent surface film. It is of interest to determine when these effects may be considered as significant.

Various workers have either discussed or investigated the factors influencing the permeability for composite reinforcements. The Carmen-Kozeny equation is frequently used to describe flow through packed beds of spherical particles and this approach has been extended to describe flow through packed beds of fibres. By the use of two correction factors, it is possible to describe flow through ideal beds of various types, with sufficient accuracy for most purposes. Although the general form of the permeability relationship is the same for flow both parallel and perpendicular to fibres in a fibre bed, the values of the correction factors will be different in each case. The first is a shape factor determined by the cross-section of the percolation passages, and the other is for the 'tortuosity' of the flow path,

Correlation between theoretical predictions and model flow experiments using idealised arrays of cylinders is reasonably good for flow both parallel and perpendicular to the cylinders, but the relationship of results on real composite systems is much less clear. Figure 7 shows the predictions of models and some key experimental results, from this and other investigation, in the form of logarithmic plots of the dimensionless quantity $\sqrt{S_x}/r_f$ against fibre volume fraction. Again it should be noted that $\sqrt{S_x} = t_{eff}$ for the experimental results reported here as proposed by the model. As has been reported previously by other workers the experimental results for flow parallel to the fibres tend to give permeabilities above the model predictions. This is shown by our experimental results and those of Reference 8. For perpendicular flow the reverse is often found to be the case.

The range of values of the effective film thickness can be identified for composite processing from Figure 7. Observing that most fibres have radii of 4-10 μm it can be seen that the extent to which velocity perturbations in real composite systems can penetrate into the bulk material is rather limited. Since most product section thicknesses lie in the range 1-5mm, this enables the region of interest in melt pultrusion for h/t_{eff} to be estimated as 50 to 25,000. So most sections can be regarded as effectively infinitely thick when modelling surface interactions. The film thickness can be seen to decrease rapidly with an increase in fibre volume fraction, particularly in the wipe off die. The relevance of this is that the shear stresses increase severely with any increase in fibre volume fraction, resulting in the characteristic decay distance becoming shorter.

CONCLUSIONS

1. Darcy's law for the flow of fluids in porous media has been extended to include the longer range viscous forces which result from externally applied shear flows as occur in thermoplastic melt pultrusion.
2. A method for measuring permeability under processing conditions in both stages of the melt pultrusion process has been developed and used. The results correlate strongly with previous investigations using an alternative method of measurement.
3. The characteristic distance in which the surface velocity perturbation decays into the bulk of the bed is equal to the square root of the Darcy permeability.
4. The characteristic decay distance likely to be encountered in most composite systems allows the process to be modelled using a simple exponential decay solution.
5. The characteristic decay distance decreases rapidly with an increase in fibre volume fraction due to a large increase in the shear stresses at the die interface.

ACKNOWLEDGEMENTS

The composite impregnation work at Newcastle upon Tyne University is sponsored by EPSRC and Shell KSLA. The authors would like to thank the donors of materials for this project; Owens Corning Fibreglass, and Uniroyal Chemicals. Further gratitude is given to Andrew Miller for performing the rheometry.

NOMENCLATURE

x,y,z	Co-ordinate axes
η	Melt viscosity
p	Pressure
v	Resin velocity vector
v_x	Fluid velocity relative to the stationary surface
f	Resin body force vector
L_D	Length of die of constant cross-section
L_P	Overall wrap length over pins
S_{xx}	Permeability in co-ordinate direction
h	Channel half-thickness
U	Velocity of the fibres relative to the stationary surface
t_{eff}	Equivalent resin film thickness
r_f	Fibre radius
V_f	Fibre volume fraction
D	Diameter of die
w	tow width

REFERENCES

1. **Bijsterbosch, H., Gaymans, R.J. and Kalisvaart, L.,** *'Pultrusion with a Nylon 6 Melt',* Proceedings of Fibre Reinforced Composites '90, I. Mech. E. and P.R.I., University of Liverpool, (April 1990).
2. **Gibson, A.G.,** *'Modification of Darcy's law to model mould interface effects in composite processing'* Composites Manufacturing, Vol. 3, No. 2, (1992), pp113-118.
3. **Brinkman, H.C.** *'A calculation of the viscous force exerted by a flowing fluid on a dense swarm of particles.'* Appl. Sci. Res, Vol. A1, (1947), pp.27-34.
4. **Williams, J.G., Morris, C.E.M. and Ennis, B.C.** *'Liquid flow through aligned fibre beds'* Polym Eng Sci, Vol. 14 (1974) pp 413-419.

5 **Drummond, J.E. and Tahir, M.I.** '*Laminar viscous flow through regular arrays of parallel solid cylinders*' Int. J. Multiphase Flow, Vol. 8, (1982), pp 193-206).

6 **Sangani, A.S. and Yao, C.** '*Transport processes in random arrays of cylinders*' Physics of Fluids, Vol. 31, (1988), pp 2435-2444.

7 **Lam, R.C. and Kardos, J.L.** '*The permeability of aligned and cross-plied fiber beds during processing of continuous fiber composites*' Proceedings of the 3rd Technical Conference, American Composites Society, Seattle, (September 1988), pp 3-11.

8 **Gutowski, T.G., Cai, Z., Bauer, S., Boucher, D., Kingery, J. and Wineman, S.** '*Consolidation experiments for laminate composites*', J. Comp Mater, Vol. 21, (1987), pp 650-669.

9 **Rudd, C.D. and Bulmer, L.** Personal Communication, University of Nottingham, (1994)

Table 1: Shear stress and film thickness results for the impregnation of glass fibre tows in a polypropylene melt when passed over seven pins at different processing speeds.

Line Speed m/min	**Shear Stress** kN/m^2	**Equivalent Film Thickness, t_{eff}** µm
1	**42.57**	**50.10**
3	**102.21**	**35.58**
5	**116.92**	**47.54**
10	**158.86**	**57.43**

Table 2. Shear stress and film thickness results for the impregnation of glass fibre tows in a polypropylene melt when passed through wipe-off dies of different cross-section at different processing speeds.

	Diameter of wipe off die mm.					
Line Speed m/min.	**3.5**	**3.0**	**2.5**	**3.5**	**3.0**	**2.5**
	Shear Stress kN/m^2			**Film Thickness** µm		
1	**3.25**	**31.05**	**67.60**	**1627.98**	**29.00**	**7.24**
3	**21.32**	**49.10**	**119.99**	**1701.43**	**38.41**	**7.80**
5	**27.41**	**49.76**	**309.29**	**181.07**	**62.53**	**2.40**
10	**31.89**	**89.99**	**455.36**	**276.56**	**43.46**	**2.41**

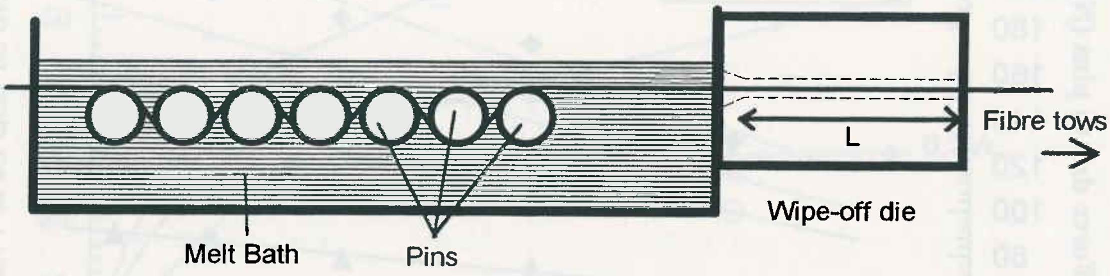

Figure 1. Melt Pultrusion by pin impregnation and wipe-off die.

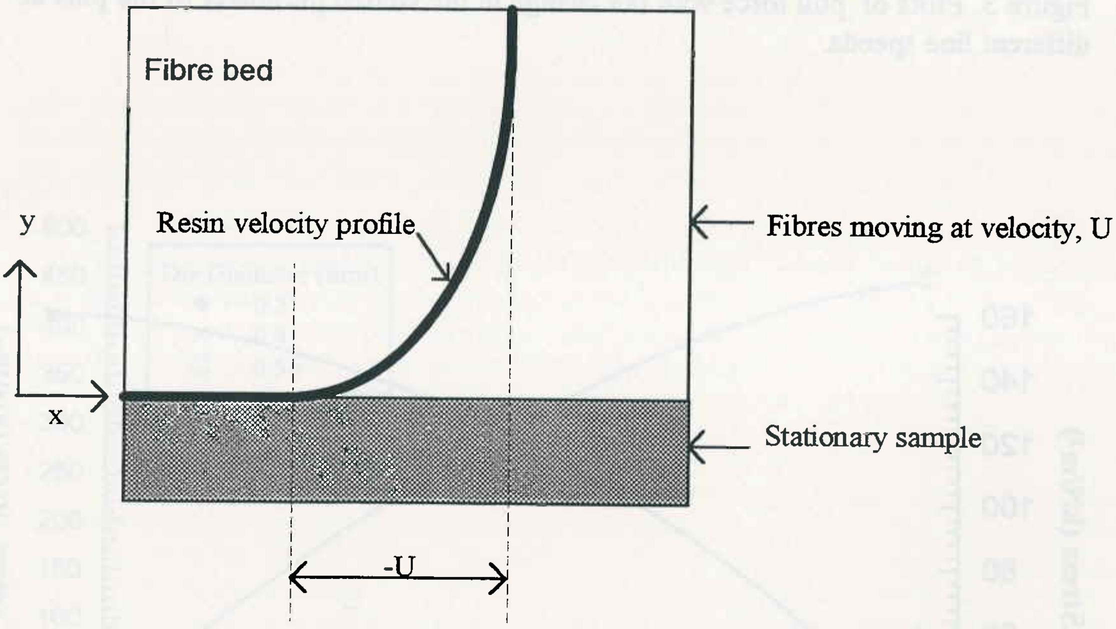

Figure 2. Velocity profile for resin flow in a fibre bed near to the interface between the bed and the pin or die wall.

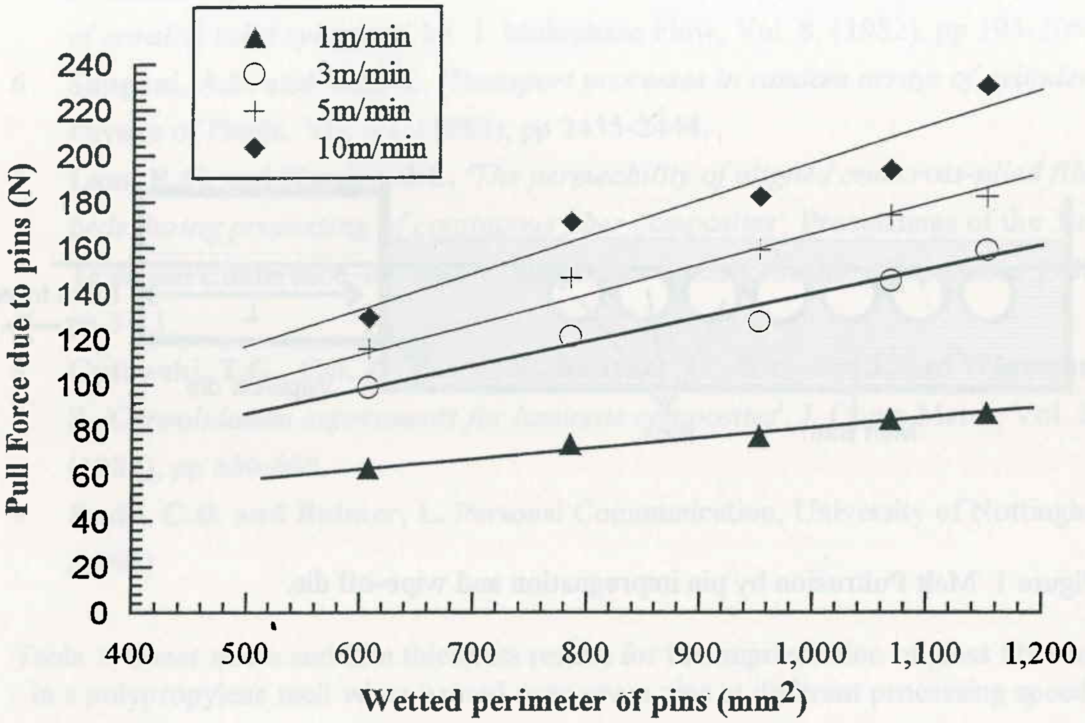

Figure 3. Plots of pull force with the change in the wetted perimeter of the pins at different line speeds.

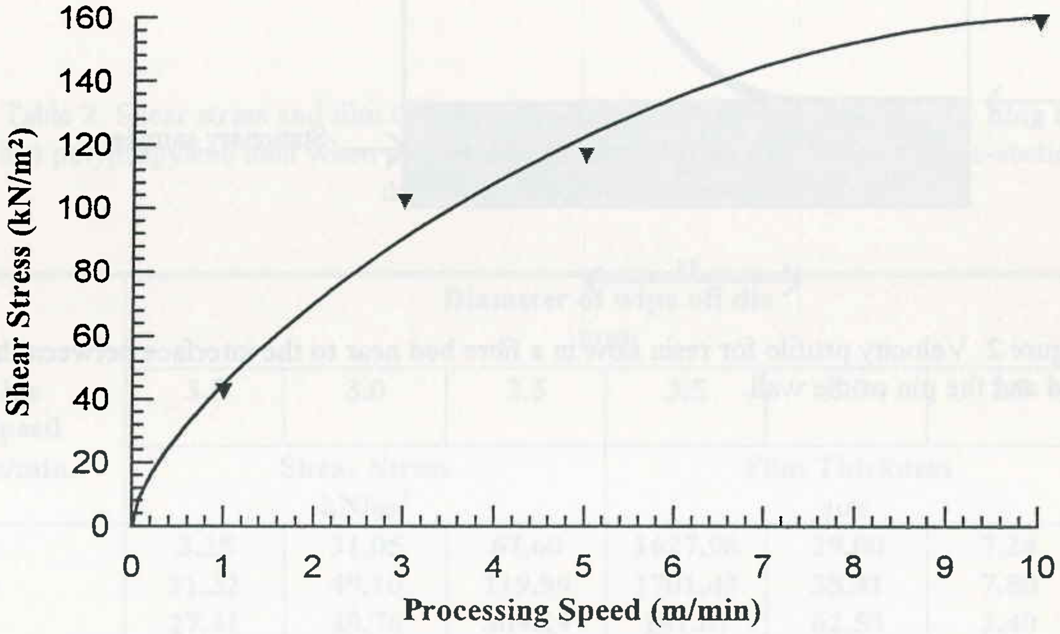

Figure 4. Relationship between shear stress at the pin surface with line speed, U for pin impregnation.

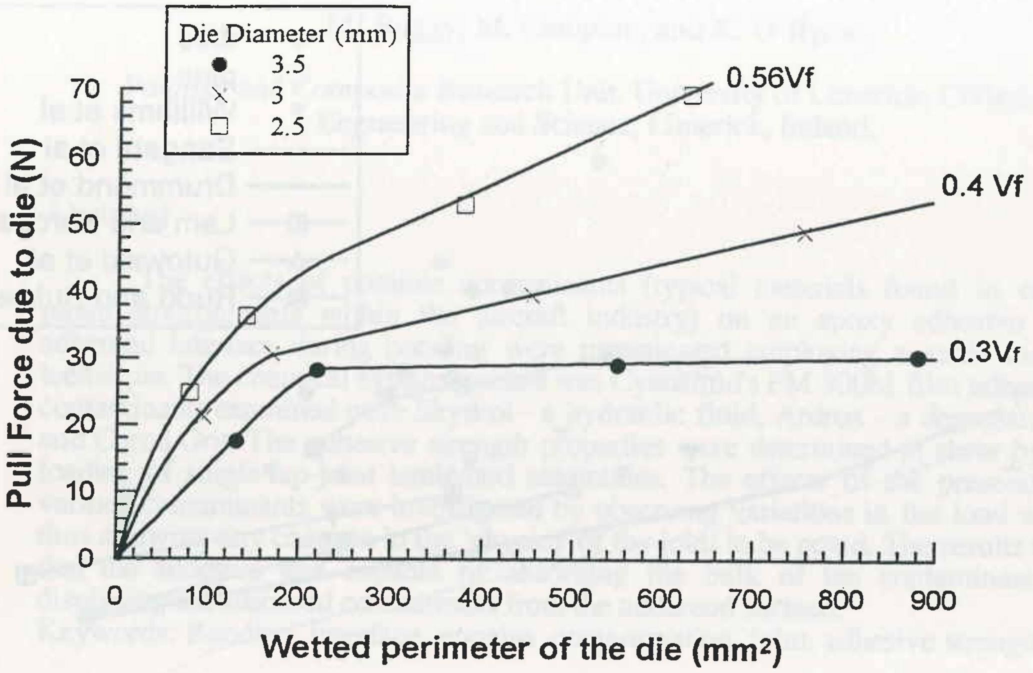

Figure 5. Variation of pull force with wetted perimeter of dies at a processing speed of 1m/min.

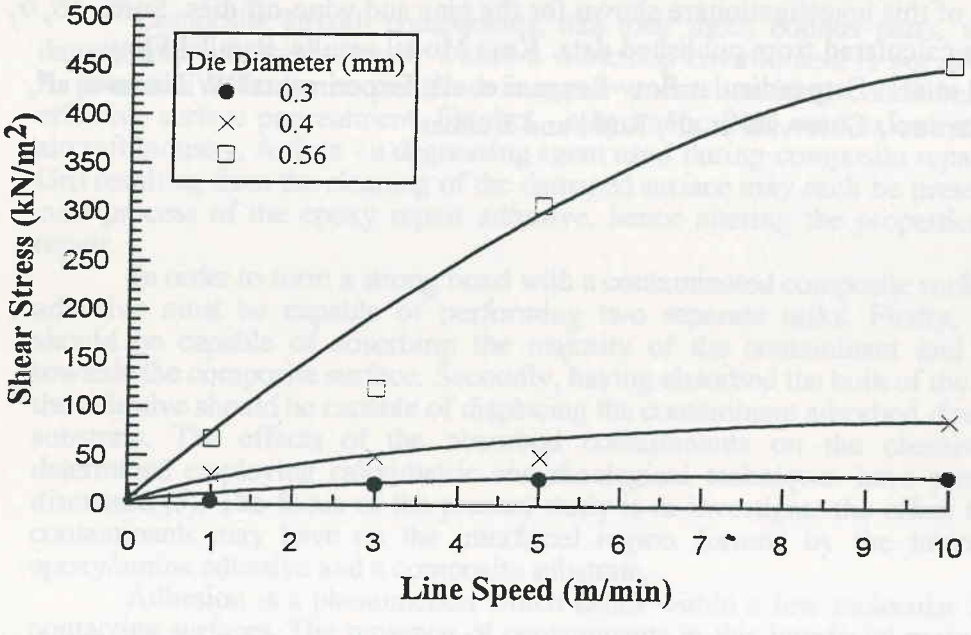

Figure 6. The relationship between shear stress at the die surface with line speed, U for wipe-off dies of three different constant cross sections.

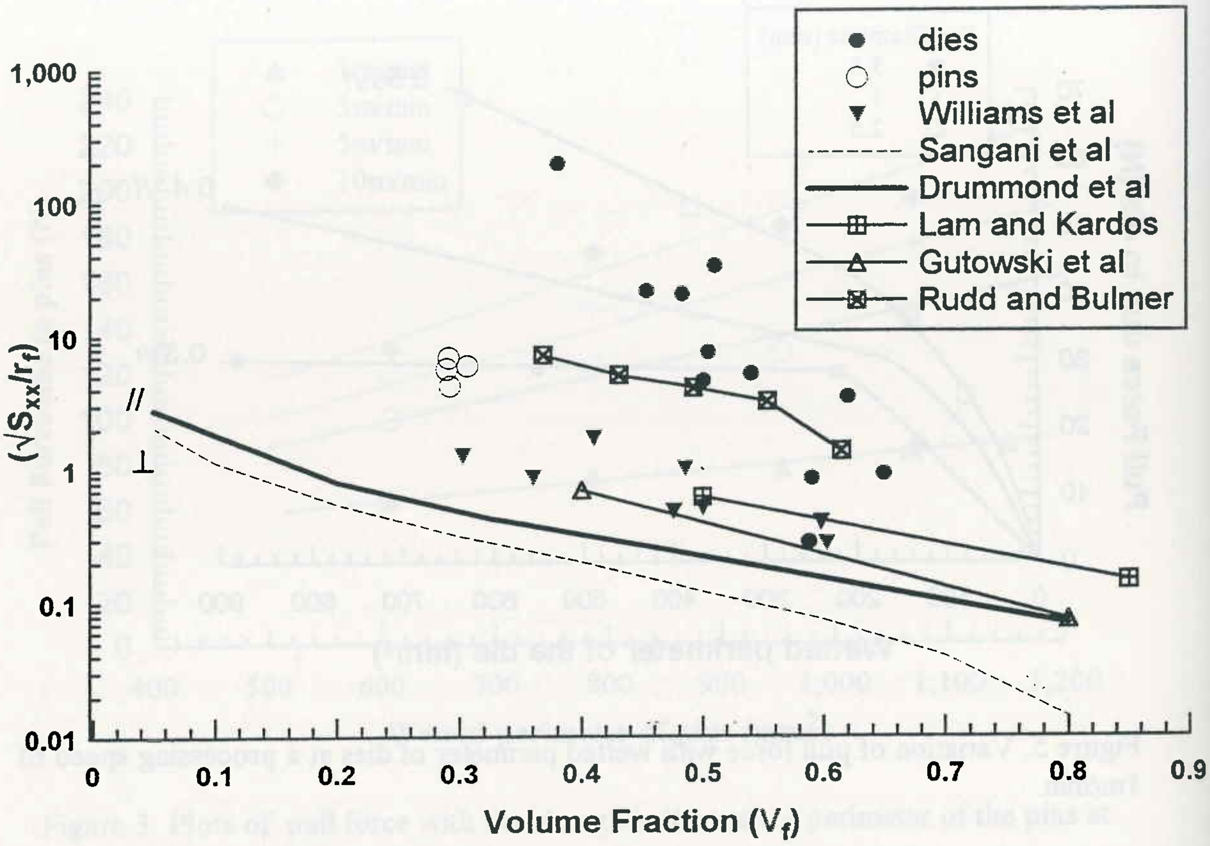

Figure 7. A comparison of results on the relationship between the square root of thepermeability/ fibre radius, $\sqrt{S_x}/r_f$ (shown on a log scale), and fibre volume fraction. The results of this investigationare shown for the pins and wipe-off dies. Some $\sqrt{S_x}/r_f$ values were calculated from published data. Key: Model results; Parallel Flow-Drummond et al[5] , Perpendicular flow-Sangani et al[6], Experimental; Williams et al[4], Lam and Kardos[7], Gutowski et al[8], Rudd and Bulmer[9]