

## **Analysis of fibre wrinkling during squeezing flows of fibre-reinforced composites**

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**Abstract.** An analysis of the stability of squeezing flows between flat plates (consolidation flows) of viscous liquids reinforced by continuous fibres is presented. The ideal linear fibre-reinforced fluid model is used to model the composite as an incompressible Newtonian fluid reinforced with inextensible fibres. The development of small fibre wrinkles initially present in the prepregged plies is analysed using linear stability theory. It is shown that when the flows are lubricated by resin rich layers, two perturbation modes are possible. In the first mode, the wrinkles are of the same form throughout the thickness of the sample while in the second mode they vary linearly with distance from the platens. In both cases the stability depends on the normal components of the applied stress. If the only traction acting in addition to hydrostatic pressure is that due to the squeezing force then the first perturbation mode is stable. This prediction is in agreement with experimental results.

### **1. Introduction**

Early models of molten fibre-reinforced polymers were developed to describe the compressive flows of sheet moulding compounds (SMC). These compounds consist of sheets of thermosetting polymer reinforced by a random array of chopped strand fibres laid in the plane of the sheets. The models were based on incompressible isotropic fluids which had either Newtonian [1] or non-Newtonian (power law) behaviour [2]. Barone and Caulk [3] described the molten layers of SMC as a transversely isotropic fluid with the axis of transverse isotropy perpendicular to the sheets. They developed a constitutive relation which was comprised of a hydrostatic reaction stress and an extra stress. The extra stress was assumed to depend on the orientation of the reinforcing strands and linearly on the rate of strain tensor. They justified this latter assumption by noting that during squeezing flows these compounds exhibited little inter-ply shearing and hence the rate of deformation was relatively small.

More recently, interest has focused on flows of thermoplastic polymers reinforced with continuous fibres. When these composites are heated until molten, the resulting fluid is also transversely isotropic but in this case the axis of transverse isotropy coincides with the fibre direction. Squeezing flows of these fluids can be used to consolidate and/or shape stacks of prepregged plies. Experimental investigations of squeezing flows between flat plates [4] which are used for consolidation of plies have found that the predominant mechanism is shearing transverse to the fibre direction. A theoretical investigation of these flows under frictionless, slip and no-slip boundary conditions has been reported [5] using the constitutive equation for 'ideal linear fibre-reinforced liquids' [6]. Similar terminology was first applied in the context of solid composites [7]; ideal models describe composites which are incompressible and reinforced by inextensible fibres.

In this paper the stability of consolidation flow is examined by using a perturbation of the basic solution for the frictionless squeezing flow of an ideal linear fibre-reinforced fluid [5].

The analysis is aimed at predicting which stress boundary conditions lead to unstable flows. This is of practical importance since such flows cause fibre wrinkling [8] and consequently reduce the compressive strength of the finished product [9]. The technique of linear stability analysis has been used recently to study steady-state flows [10, 11], but this paper is believed to contain the first analysis of a flow of an anisotropic liquid whose unperturbed solution is time dependent.

## 2. Governing equations

The equations are formulated in the rectangular cartesian coordinates  $x_i$  with the velocity components denoted by  $u_i$ . The index  $i$  takes the values 1, 2 and 3 and the usual summation convention applies to repeated indices. The fibre direction is represented by a unit vector  $\mathbf{a}$  which has components  $a_i$ .

In the continuum model being used it is assumed that the fluid is incompressible and that the fibres are inextensible. These two kinematic constraints may be expressed [7] as

$$D_{ii} = 0, \quad (1)$$

$$a_i a_j D_{ij} = 0, \quad (2)$$

where the components of the strain rate tensor are  $\mathbf{D}$  defined as

$$D_{ij} = \frac{1}{2}(\partial u_i / \partial x_j + \partial u_j / \partial x_i). \quad (3)$$

It is also assumed that the fibres are locally parallel, continuously distributed and that they convect with the fluid. Thus, a section of fibre always remains adjacent to the same fluid element during a flow and this condition, together with the constraint of fibre inextensibility yields [7]

$$\partial a_i / \partial t + u_j \partial a_i / \partial x_j = a_j \partial u_i / \partial x_j, \quad (4)$$

where  $t$  denotes time.

To complete the description of the ideal linear fibre-reinforced fluid a constitutive equation for the stress is required. In general the stress  $\boldsymbol{\sigma}$  may be written as the sum of an indeterminate reaction stress  $\boldsymbol{\sigma}^R$  and a determinate extra stress  $\boldsymbol{\sigma}^E$ . The reaction stress arises as a consequence of the imposed kinematic constraints and for ideal fibre-reinforced materials it takes the form [7]

$$\sigma_{ij}^R = -\Pi \delta_{ij} + T a_i a_j, \quad (5)$$

where  $\delta_{ij}$  denotes the Kronecker delta. The first term denotes an hydrostatic pressure  $\Pi$  and the second denotes a fibre tension  $T$  which arises from the assumption of fibre inextensibility. This assumption is applicable to materials in which the resistance to motion in the fibre direction far exceeds that in other deformation modes. It is appropriate for analysis of squeezing flows since experimental evidence [4] indicates that comparatively little flow occurs in the fibre direction.

The prefix 'linear' in the nomenclature 'ideal linear fibre-reinforced liquid' refers to the assumption that the extra stress  $\boldsymbol{\sigma}^E$  depends linearly on the strain rate tensor  $\mathbf{D}$ . There is

some experimental evidence [12] to support this assumption of a linear dependence of extra stress on strain rate at least for the slow flows. The extra stress also depends on the fibre direction and because the sense of the fibres is inconsequential (the stress is unchanged if the direction of the fibres is reversed) it is sufficient to assume a dependence on the product  $a_i a_j$ . The form of the extra stress can be formally derived from consideration of tensor functions of  $D_{ij}$  and  $a_i a_j$  which are invariant under rigid body rotations. This gives [6]

$$\sigma_{ij}^E = 2\eta_T D_{ij} + 2(\eta_L - \eta_T)(a_i a_k D_{kj} + D_{ik} a_k a_j). \quad (6)$$

The transverse viscosity  $\eta_T$  and longitudinal viscosity  $\eta_L$  relate to shearing perpendicular to and along the fibre direction, respectively. Typical values for a composite having reinforcement of 35% by volume are  $1.3 \times 10^3$  Pa s and  $2.0 \times 10^3$  Pa s [13]. There is some evidence both experimental [13] and theoretical [14] to suggest that the relative size of these viscosities depends on the proportion of reinforcing fibres, with a higher concentration of fibres leading to a larger value of the transverse viscosity relative to the longitudinal viscosity.

As noted above the reaction stress is arbitrary in the sense that it is not defined by a constitutive relation. Rather the pressure  $\Pi$  and tension  $T$  are determined from the equations of motion which in the absence of body forces take the form

$$\partial \sigma_{ij} / \partial x_j = \rho(\partial u_i / \partial t + u_j \partial u_i / \partial x_j), \quad (7)$$

where  $\rho$  denotes the density of the composite. As in the case of *isotropic* viscous fluids the character of the flow depends on the relative magnitude of the inertial and viscous terms. If  $U$  is a characteristic velocity and  $L$  is a characteristic length scale then transverse and longitudinal Reynolds numbers may be defined as

$$R_T = (UL\rho) / \eta_T \quad \text{and} \quad R_L = (UL\rho) / \eta_L. \quad (8)$$

For the flows under discussion typical values of these parameters are

$$U = 5 \text{ mm s}^{-1}, \quad L = 5 \text{ mm}, \quad \rho = 1500 \text{ kg m}^{-3},$$

and using the viscosities cited above gives

$$R_T = 2.88 \times 10^{-5} \quad \text{and} \quad R_L = 1.88 \times 10^{-5}. \quad (9)$$

Thus, the inertial terms may be neglected and the slow flow equations

$$\partial \sigma_{ij} / \partial x_j = 0, \quad (10)$$

are suitable for describing these flows. This assumption is used for the remainder of this paper.

To complete the specification of the problem boundary conditions must be considered. For viscous fluids the adherence condition is the almost universally accepted condition at solid boundaries and it would be natural to extend this condition to the flow of anisotropic fluids. However, there is experimental evidence [3, 4] to suggest that slip does occur during flows of some composite materials. The most likely explanation of this is the presence of thin resin rich layers which lubricate the flow when formed between the composite and the platen surfaces. In addition to this 'natural' lubrication, artificial lubricants in the form of chemical

release agents are often applied to the platen surfaces to ensure that the composite can be removed easily after consolidation. It has been suggested [4] that these lubricated flows can be modelled by applying slip boundary conditions in which the shear stress at a solid boundary is proportional to the relative velocity between the fluid and boundary. In cases where the boundary is frictionless the shear stress is zero at the boundary.

### 3. Consolidation flows

Figure 1 illustrates the flow being modelled. At time  $t = 0$  the two platens are at rest in the planes  $x_2 = H_0$  and  $x_2 = 0$  with the viscous ideal reinforced liquid lying between the platens and the fibres aligned parallel to the  $x_3$ -axis. For  $t > 0$  the upper platen is pushed down by a force acting in the negative  $x_2$ -direction (of magnitude  $F(t)$  per unit length of the  $x_3$ -axis), while the lower platen remains fixed. The dimension of the fluid in the  $x_1$ -direction is denoted by  $2L(t)$  at any time  $t$  and  $2L_0$  denotes its initial ( $t = 0$ ) value.

If both platens are perfectly lubricated then the governing equations admit the homogeneous, time dependent solution [5]

$$\begin{aligned} \mathbf{u} &= (-Bx_1, Bx_2, 0), & \mathbf{a} &= (0, 0, 1), \\ \sigma_{11} &= -\Pi - 2\eta_T B, & \sigma_{22} &= -\Pi + 2\eta_T B, & \sigma_{33} &= -\Pi + T, \\ \sigma_{12} &= \sigma_{13} = \sigma_{23} = 0. \end{aligned} \tag{11}$$

where

$$B(t) = \dot{H}/H, \quad L(t) = L_0 H_0 / H(t).$$

The superposed dot denotes differentiation with respect to time. The expression for  $L(t)$  follows directly from incompressibility.

The solution for the hydrostatic pressure is obtained by satisfying the traction boundary conditions at the edges  $x_1 = \pm L(t)$ . If these edges are traction free then

$$\Pi = -2\eta_T B. \tag{12}$$

If it is further assumed that there is zero traction on the edges ( $x_3 = \text{constant}$ ) normal to the fibre direction then the fibre tension  $T$  is

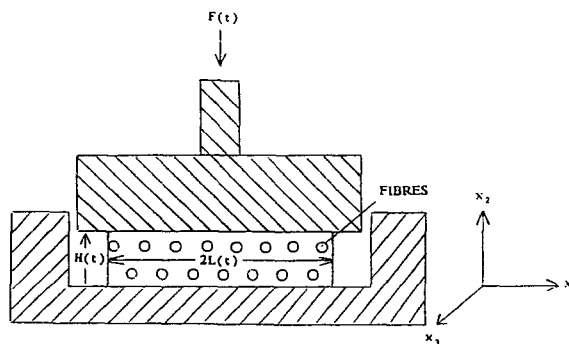


Fig. 1. Consolidation flow of a fibre-reinforced composite.

$$T = -2\eta_T B(t) = -2\eta_T \dot{H}/H. \tag{13}$$

From this result it follows that squeezing flows (in which  $\dot{H}$  is negative) induce a positive value for  $T$ . Although this result has been obtained for the special case of zero applied tractions acting on the edges normal to the  $x_1$  and  $x_3$ -axes it remains valid for any values of these tractions which are equal.

The separation  $H(t)$  of the platens can be related to the applied force  $F(t)$  acting on the platen surface  $x_2 = H(t)$  by integration of the stress component  $\sigma_{22}$ . Using the solution for the pressure in equation (12) this gives

$$H(t) = \frac{8\eta_T L_0 H_0}{8\eta_T L_0 + \int_0^t F(\tau) d\tau}. \tag{14}$$

#### 4. Stability analysis

It is well known that in the preimpregnated plies (prepregs) which are the precursor material used in squeezing flows the fibres are not perfectly collimated; off-axial deviations of up to  $6^\circ$  have been reported [15]. The ensuing analysis examines the effect of frictionless squeezing flows on these imperfectly collimated fibres. It is particularly important to determine whether such flows tend to improve the alignment or the fibres of cause greater misalignment since poorly collimated fibres lead to a reduction in strength of the finished product [9].

It is assumed that the magnitude of the initial fibre misalignments is small and so can be represented by a small dimensionless parameter  $\epsilon$ . The variables can then be expanded in perturbation series of the form:

$$\begin{aligned} \mathbf{u} &= \mathbf{u}^0(x_i, t) + \epsilon \mathbf{u}^1(x_i, t) + O(\epsilon^2), \\ \mathbf{a} &= \mathbf{a}^0(x_i, t) + \epsilon \mathbf{a}^1(x_i, t) + O(\epsilon^2), \\ \Pi &= \Pi^0(x_i, t) + \epsilon \Pi^1(x_i, t) + O(\epsilon^2), \\ T &= T^0(x_i, t) + \epsilon T^1(x_i, t) + O(\epsilon^2). \end{aligned} \tag{15}$$

The first terms in the series may be regarded as the solutions for perfectly collimated fibres in which  $\epsilon = 0$ ; such solutions are given in equation (11). The second term in the series expansion of the fibre direction vector represents a small perturbation in the fibre direction. This in turn causes perturbations to the basic velocity, pressure and tension fields. Terms of second order and higher in  $\epsilon$  are neglected.

It follows from the definition of  $\mathbf{a}$  as a unit vector and the solution for  $\mathbf{a}^0$  given in equation (11) that  $a_3^1$  is zero. Also since the fibres are assumed to convect with the fluid no first order perturbation in velocity occurs in this direction and so  $u_3^1 = 0$ . The squeezing flow inhibits the development of any perturbations in the  $x_2$ -direction and so solutions are sought for which

$$u_2^1 = a_2^1 = 0. \tag{16}$$

Then the only non-zero first-order terms are  $u_1^1$  and  $a_1^1$ . The incompressibility condition requires that  $u_1^1$  must be independent of  $x_1$ . Furthermore, the fibre perturbation component  $a_1^1$  is chosen to be independent of  $x_1$ .

Real samples often exhibit a wavelike disturbance which can be approximately modelled

as a sinusoidal variation in the direction of the unperturbed fibres [9]. Following this approach non-zero perturbations are sought of the form:

$$\begin{aligned} u_1^1(x_2, x_3, t) &= u_1^*(x_2, t) \sin(kx_3), \\ a_1^1(x_2, x_3, t) &= a_1^*(x_2, t) \cos(kx_3), \\ \Pi^1(x_2, x_3, t) &= \Pi^*(x_2, t) \cos(kx_3), \\ T^1(x_2, x_3, t) &= T^*(x_2, t) \cos(kx_3). \end{aligned} \quad (17)$$

The wavelength of the disturbances is  $2\pi/k$  where  $k$  is a real number.

The governing equations for the first order perturbations are obtained by substituting the series described in equations (15) and (17) into the convection equations (4) and the slow flow equations of motion (10), and then equating terms of  $O(\epsilon)$ . The only non-trivial first order convection equation provides a relation between the components  $a_1^*$  and  $u_1^*$  of the form

$$\partial a_1^* / \partial t + B \partial(x_2 a_1^*) / \partial x_2 = k u_1^*. \quad (18)$$

The first of the equations of motion (10) leads to a second relation between these components

$$\eta_T \partial^2 u_1^* / \partial x_2^2 - k^2 \eta_L u_1^* - k \{ T^0 - 2B(\eta_L - \eta_T) \} a_1^* = 0, \quad (19)$$

while the second and third equations of motion are satisfied by choosing

$$\Pi^* = T^* = f(t), \quad (20)$$

where  $f(t)$  is an arbitrary function of time.

Equation (18) can be used to eliminate  $u_1^*$  from equation (19) to give the third order partial differential equation for  $a_1^*$

$$\left\{ \eta_T \frac{\partial^2}{\partial x_2^2} - k^2 \eta_L \right\} \left\{ \frac{\partial a_1^*}{\partial t} + B \frac{\partial(x_2 a_1^*)}{\partial x_2} \right\} - \{ T^0 - 2B(\eta_L - \eta_T) \} k^2 a_1^* = 0. \quad (21)$$

The time dependent behaviour of the solutions of this equation provides information about the growth-decay of the first order fibre perturbations. Two solutions are now considered.

#### 4.1. *Solution independent of depth*

Equation (21) has a time dependent solution  $a_1^*(t)$  which satisfies the equation

$$\eta_L da_1^* / dt + \{ T^0 + B(2\eta_T - \eta_L) \} a_1^* = 0. \quad (22)$$

Integration of this equation yields the solution

$$a_1^*(t) = P(t) = C_1 I_1(t), \quad (23)$$

where  $C_1$  is a constant and the integral  $I_1(t)$  is

$$I_1(t) = (-1/\eta_L) \int_0^t \{T^0 + B(2\eta_T - \eta_L)\} d\tau . \quad (24)$$

This integral can be rewritten in terms of the unperturbed stress components by making use of solution (11) to give

$$I_1(t) = \frac{1}{4\eta_L\eta_T} \int_0^t \{4\eta_T(\sigma_{11}^0 - \sigma_{33}^0) + \eta_L(\sigma_{22}^0 - \sigma_{11}^0)\} d\tau . \quad (25)$$

The non-zero velocity component  $u_1^*$  is determined directly from equation (18) as

$$u_1^* = (C_1/k\eta_L)\{2B(\eta_L - \eta_T) - T^0\} \exp\{I_1(t)\} . \quad (26)$$

It then follows from equations (17) and (20) that the full solution is:

$$\begin{aligned} a_1^1 &= C_1 \cos(kx_3) \exp\{I_1(t)\} , & a_2^1 &= a_3^1 = 0 , \\ u_1^1 &= (C_1/k\eta_L) \sin(kx_3) \{2B(\eta_L - \eta_T) - T^0\} \exp\{I_1(t)\} , & u_2^1 &= u_3^1 = 0 , \\ \Pi^1 &= T^1 = f(t) \cos(kx_3) . \end{aligned} \quad (27)$$

It is clear that the stability of the flow depends on the integral  $I_1(t)$  defined in equations (24) and (25). It follows from the latter equation that varying the zero order hydrostatic pressure has no effect on the stability of the flow. Rather the growth or decay of the sinusoidal perturbations depends entirely on two normal stress differences which may be conveniently defined as

$$N_1 = \sigma_{11}^0 = \sigma_{33}^0 , \quad N_2 = \sigma_{22}^0 - \sigma_{11}^0 . \quad (28)$$

Using these definitions it follows that if

$$4\eta_T N_1 + \eta_L N_2 < 0 \quad \text{for all } t > 0 ,$$

then the flow is stable, and if

$$4\eta_T N_1 + \eta_L N_2 > 0 \quad \text{for all } t > 0 ,$$

then the flow is unstable.

If the value of the applied normal stress component  $\sigma_{33}^0$  which acts in the fibre direction is increased and the value of the normal stress component  $\sigma_{11}^0$  is kept fixed, then the normal stress difference  $N_1$  will decrease and the flow will become more stable. This result is in agreement with intuitive reasoning which suggests that the application of increasing tensile forces in the fibre direction should tend to reduce the fibre wrinkling.

Now consider the effect of the normal stress difference  $N_2$ . It follows from equations (28) and (11) that

$$N_2 = \sigma_{22}^0 - \sigma_{11}^0 = 4\eta_T B = 4\eta_T \dot{H}/H . \quad (29)$$

Thus, if all other factors are equal, wrinkled fibres in a sample undergoing consolidation with a high squeezing rate (that is a large negative value of  $\dot{H}$ ) will be straightened more quickly than those in a sample which is consolidated with a low squeezing rate.

In consolidation flows in which the only non-zero applied force is acting in the  $x_2$ -direction

the value of the unperturbed tension  $T^0$  is given by equation (13). It then follows from equation (24) that the integral  $I_1(t)$  becomes

$$I_1(t) = \int_0^t B \, d\tau = \int_0^t (\dot{H}/H) \, d\tau = \ln\{H(t)/H_0\} . \quad (30)$$

In this case the amplitude of the fibre wrinkles exhibits exactly the same time dependent behaviour as the platen separation  $H(t)$ . Thus, closing the platens quickly will cause the amplitude of the fibre wrinkled to decrease quickly. If a squeezing force of smaller magnitude is applied the platens will close more slowly and hence the fibres will straighten at a slower rate. However, if two initially identical samples are squeezed to the same final dimensions then the extent of the fibre wrinkling in the consolidated samples will be the same.

Experiments carried out by Jones and Roberts [16] using a model composite are in qualitative agreement with the predictions of the depth independent solution (27). Their experiments were carried out under atmospheric pressure with a squeezing force applied in the  $x_2$ -direction. It was found that when the platens were lubricated, artificially wrinkled fibres which were present throughout the sample all straightened out.

#### 4.2. Solution linear in $x_2$

Equation (21) also has a solution which is linear in  $x_2$  of the form

$$a_1^* = x_2 Q(t) . \quad (31)$$

Substitution of this solution into equation (21) yields the first order differential equation for  $Q(t)$

$$\eta_L \, dQ/dt + (T^0 + 2B\eta_T)Q = 0 , \quad (32)$$

which can be integrated to give

$$Q(t) = C_2 \exp\{I_2(t)\} , \quad (33)$$

where  $C_2$  is a constant and the integral  $I_2(t)$  is

$$\begin{aligned} I_2(t) &= \frac{-1}{\eta_L} \int_0^t \{T^0 + 2B\eta_T\} \, d\tau , \\ &= \frac{1}{\eta_L} \int_0^t (\sigma_{11}^0 - \sigma_{33}^0) \, d\tau = \frac{1}{\eta_L} \int_0^t N_1 \, d\tau . \end{aligned} \quad (34)$$

The full solution for this mode of fibre wrinkling is:

$$\begin{aligned} a_1^1 &= x_2 C_2 \cos(kx_3) \exp\{I_2(t)\} , \quad a_2^1 = a_3^1 = 0 , \\ u_1^1 &= \frac{x_2 C_2 \sin(kx_3) \{-T^0 + 2B(\eta_L - \eta_T)\} \exp\{I_2(t)\}}{k\eta_L} , \quad u_2^1 = u_3^1 = 0 , \\ \Pi^1 &= T^1 = f(t) \cos(kx_3) . \end{aligned} \quad (35)$$



The function  $f(t)$  is arbitrary.

In this case the stability of the flow depends only on the normal stress difference  $N_1$  which is defined in equation (28). The stability conditions for this mode are that if

$$N_1 < 0 \quad \text{for all } t > 0,$$

then the flow is stable, and that if

$$N_1 > 0 \quad \text{for all } t > 0,$$

then the flow is unstable.

In the case when the normal stresses  $\sigma_{11}^0$  and  $\sigma_{33}^0$  are equal, as happens when the only applied force acts in the  $x_2$ -direction, the stress difference  $N_1$  becomes zero and the behaviour of the perturbations is time independent. In such flows any wrinkles which are initially present in the plies remain unaltered by the consolidation process. It is noted that the stability criteria are independent of the squeezing force, that is the force applied in the  $x_2$ -direction.

The solution (35) which is linear in  $x_1$  describes the stability behaviour of a sample which initially has an inhomogeneity which is linear in  $x_2$ . The magnitude of this inhomogeneity need only be small, that is, of order epsilon. Such a case could be realised in practice if the laminate is assembled from several different plies of prepregged material since different plies could possess different degrees of fibre wrinkling due to variability in their manufacture.

## 5. Conclusions

Two solutions have been found for the first order perturbation equations governing the frictionless squeezing flow of ideal fibre-reinforced liquids. In the first mode the sinusoidal fibre wrinkles are time dependent and independent of distance from the platens. In this mode the stability depends on two normal stress differences. If the traction applied in the direction of the reinforcing fibres exceeds that which acts in the direction of the elongational flow, and if this in turn is greater than the traction acting on the platen surface, then the flow is always stable. The special case, widely used in practical applications, in which the squeezing is caused by an applied force acting only in the vertical ( $x_2$ ) direction has been found to straighten the fibres irrespective of what magnitude of squeezing force is applied. Experimental results are in qualitative agreement with the predictions of this perturbation mode.

The second mode describes the behaviour of a sample which is initially inhomogeneous. If the inhomogeneity varies linearly in the vertical direction the stability of the flow depends only on the difference between the two normal stress components in the plane of the plies.

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