Insights into complex rheological behaviour of carbon fibre/PEEK from a novel numerical methodology incorporating fibre friction and melt viscosity

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Abstract:

A recent rheological study of carbon-fibre-reinforced PEEK (CF/PEEK) demonstrated highly complex behaviour, involving phenomenological differences at low and high strain rates. To explain the behaviour, it was hypothesised that CF/PEEK responds as a yield-stress fluid at low strain rates, with boundary-lubricated, fibre-fibre friction determining the viscosity, and as a viscous fluid at high strain rates, with polymer melt viscosity dominating the response. In this paper, a novel finite-element methodology, incorporating fibre friction and melt viscosity in the same model, is employed to study this hypothesis. Two-fibre models investigate how fibre friction and melt viscosity combine to produce an overall composite viscosity. Representative-volume-element (RVE) models examine multi-fibre/melt response, and demonstrate that inclusion of fibre friction produces the observed yield-stress behaviour at low strain rates, and viscous behaviour at high strain rates. Another phenomenon which affects rheological measurements of such composites is shear banding in the sample, which occurs in the yield-stress regime. This effect is demonstrated in the models, and analysis of load transfer between fibres and melt explains how it arises, and how it leads to diminished values of measured viscosity. The results pave the way for improved process models for high-throughput manufacturing processes such as Automated Tape Placement.

Keywords

Carbon fibres; Thermoplastic Resin; Viscosity; Finite Element Analysis (FEA);

1. Introduction

Having recently purchased a laser-heated Automated Tape Placement (ATP) system, the Irish Composites Centre (IComp) is investigating ATP processing of carbon-fibre-reinforced poly-ether-ether-ketone (CF/PEEK) [1, 2], a high-performance thermoplastic composite, qualified for the aerospace industry. In ATP, it is generally assumed, [3], that no-slip boundary conditions exist between the roller and the top of the pre-preg tape, and

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between the bottom of the tape and the previously consolidated layer. These conditions lead to squeeze flow, with transverse shear being the dominant deformation mode [3]. Thus, transverse-shear viscosity of CF/PEEK melt is considered a key parameter for models of the process.

Although CF/PEEK has been extensively characterised, the literature reveals large variations in reported values of transverse-shear viscosity [4-7], with no satisfactory explanation for why this is. For example, as can be seen in Fig. 1, the data of Groves et al. [5] differs from that of O'Bradaigh et al. [6] by two orders of magnitude at a strain rate of 1 s^{-1}. This lack of clarity makes choosing values for ATP process models of CF/PEEK very problematic. To investigate the reasons for these wide discrepancies in the literature, we have undertaken an experimental and numerical study of CF/PEEK under processing conditions.

In the experimental study [8], we performed detailed measurements on Suprem™ CF/PEEK melt using a novel off-centred, balanced, oscillatory rheometry technique, on single plies to avoid interplay slip. Some of our findings confirmed previous studies, e.g. at low strain rates (10^{-3} s^{-1} to ~10 s^{-1}), the transverse-shear viscosity is shear-thinning (unlike PEEK, which is Newtonian up to strain rates of ~10 s^{-1}). However, some of our other results had not been reported before and were quite surprising. For example, frequency sweeps revealed a non-linear dependence on the pressure applied to the sample by the rheometer plates, with the viscosity measured at pressures above 156 kPa being an order of magnitude higher than those at pressures below 104 kPa. When compared to previous results (see Fig. 1), it was interesting to note that the low pressure data matched the results in [4] and [5], for which loading conditions approximated simple shear, while the high pressure data was closer to that of [6] and [7], for which combined shear-compression was applied. Other surprising results were that viscosity also increased substantially during constant-frequency oscillatory tests over just a few minutes, and *increased* with increasing temperature. These non-intuitive results, together with the wide variations in values in the literature, prompted the present numerical investigation.

Several studies have investigated topics of relevance to CF/PEEK behaviour, e.g. compressive behaviour of dry fibrous materials [9] or fibre preforms [10], including friction at contact points [11] or resistance to compression due to reverse bending of initially wavy fibres, pinned by contacts with other fibres [12]. In suspensions, the effects of numerous fibre-fibre contacts [13], time-dependent resistance to compression caused by hydrodynamic behaviour of the melt [7, 14] and increased resistance to deformation, caused by high strain rate of the melt as it is sheared between fibres [15-18], have also been examined. However, none of these studies gives a fully satisfactory explanation of available CF/PEEK data. In [8] we postulated that the behaviour seen in our experiments, and in the literature, could all be explained by considering CF/PEEK melt to be a "yield-stress fluid". Yield-stress fluids generally contain solid particles, and at low shear rates, frictional interactions between particles create a network structure that imparts a solid-like, yield-stress behaviour. For example, a frictional interaction between discontinuous fibres in a polypropylene melt, occurring at fibre crossing points, was proposed to explain yield-stress behaviour in that material [19, 20]. When the applied stress overcomes the yield stress, the network structure breaks up, and the material flows. Hence, they are often referred to as "viscoplastic materials" [21]. In CF/PEEK, it is proposed that at low shear rates, polymer flow resistance is low, and friction

between fibres determines the material response. This leads to a yield stress, τ_y, below which no fibres move, which results in an effective viscosity that decreases at a constant rate with increasing shear rate, i.e. power-law, shear-thinning behaviour. This is exactly the type of behaviour seen for CF/PEEK at the strain rates tested to date (< ~10 s^{-1}). At higher strain rates, the melt resistance will be higher, and should eventually dominate. The material is then expected to behave as a viscous fluid, with a viscosity higher than PEEK, due to the elevated polymer shear rate in small inter-fibre spaces [15]. The viscosity should eventually drop off at high enough strain rates, at which PEEK is shear-thinning.

An initial material configuration is envisaged, in which, at any given cross-section of the pre-preg, fibre waviness and tow twist result in most, if not all, fibres having some regions of their surface close enough to other fibres to be in boundary-lubricated contact, in which a layer of polymer melt may adhere to, or be absorbed into the gaps between the rough fibre surfaces. This would result in frictional interaction, but with a lower coefficient of friction than for dry friction [22]. As suggested by Barnes and Cogswell [23], twisting of fibres around each other, coupled with fibre inextensibility, results in them being forced together with relatively small normal forces in the as-produced pre-preg. According to tribology theory, boundary-lubricated friction occurs at low shear rates. At higher shear rates, increased fluid pressure could cause fibres to be pushed apart, leading to mixed, or hydrodynamically-lubricated friction, for which frictional resistance is much less [24]. With this conceptual model of material behaviour, the fibre surface area in contact will increase over time under external pressure, explaining the upwards drift in CF/PEEK viscosity over time seen in [8]. The increase in fibre contact area will be facilitated by a lower viscosity polymer, explaining the counter-intuitive increase in CF/PEEK viscosity with temperature, also seen in [8].

To explain the higher viscosity found at pressures above 156 kPa, than at pressures below 104 kPa in [8], it is proposed that shear banding occurs at low pressure, in which only the layer or layers of fibres closest to the moving platen move. Shear banding is a characteristic behaviour of yield-stress materials. Møller et al. [25], have shown that glassy, yield-stress materials, loaded in shear at low strain rates, deform in only a thin band near the shearing surface, while at high strain rates, no banding is observed. A re-examination of data from our laboratory provides evidence that shear banding can also occur in CF/PEEK melt. Data from Stanley and Mallon,[4] which was for a continuous, simple-shear test at a low constant strain-rate (0.01 s^{-1}), under low applied pressure, are re-plotted in Fig. 2. It can be seen that shear viscosity more than doubled as shear strain increased from 0 to 30%. The most likely explanation for this increase is that it is due to shear banding, in a flow regime dominated by fibre friction, which occurs as follows. Initially, only layers of fibres closest to the moving platen displace, with resistance coming primarily from fibre-fibre friction between this and the next layer. However, as the applied strain increases, fibre twisting and entanglement, as depicted by Barnes and Cogswell,[23] leads to successive layers being forced into motion, resulting in increased frictional resistance. This translates into the increase in viscosity with strain magnitude, observed in Fig. 2. The image of the side of the specimen in Fig. 2 and the stepped appearance of the data supports the shear banding hypothesis.

To bolster these hypotheses concerning CF/PEEK behaviour and explore them further, a novel finite-

element (FE) methodology, incorporating fibre friction and melt viscosity in the same model, is utilised here. The non-linear, implicit FE code, Abaqus/Standard® [26] is used throughout. A model for PEEK melt, robust enough to handle large strains without numerical problems is first developed. Two-fibre models are then used to examine the combined effects of fibre friction and melt viscosity on the overall composite viscosity. Representative-volume-element (RVE) models simulate Suprem™ CF/PEEK material, which features 5.2 μm diameter Hexcel® IM7 carbon fibres in Victrex™ PEEK. These models examine multi-fibre/melt response, with and without fibre friction and results are compared with experiments. Detailed analysis of pressure and skin-friction drag forces applied by the melt to the fibres is used to understand the means by which shear banding can occur, and how it leads to diminished values of measured viscosity. The range of shear strain rates applied in the experiments, together with higher shear strain rates, applicable to ATP consolidation, are explored.

2. Modelling Methodologies

2.1 Material model for PEEK melt

A robust model for the deviatoric behaviour of PEEK melt, free from numerical problems over a large range of strain rates (0.001 s^{-1} to 1000 s^{-1}) was a vital pre-requisite for this work. Experimental data from [8, 27] indicate that PEEK shear viscosity, η, at 380˚C, exhibits a transition from Newtonian to shear-thinning behaviour at a strain rate of ~10 s^{-1}. It also exhibits a large linear-viscoelastic region, over which, shear viscosity is constant with increasing strain. Here, PEEK is represented by a hyper-viscoelastic model. An Arruda-Boyce [28] hyperelastic potential describes the solid-like, elastic behaviour, while a Parallel Rheological Framework (PRF) [26, 29] represents the fluid-like, time-dependent, creep behaviour of the melt as a series of springs and dashpots. A four-network PRF was employed that utilises a power-law, strain-hardening model of the form:

$$\dot{\bar{\varepsilon}}^{cr} = \left(A\tilde{q}^{n} \left[(m+1)\bar{\varepsilon}^{cr} \right]^{m} \right)^{\frac{1}{m+1}} \tag{0}$$

where $\bar{\varepsilon}^{cr}$ is the equivalent creep strain, $\dot{\bar{\varepsilon}}^{cr}$ is the equivalent creep strain rate, $\tilde{q}$ is the equivalent deviatoric Kirchhoff stress, and A, m and n are material parameters. For each network, this non-linear, viscoelastic model was reduced to linear-viscoelastic behaviour by specifying $m = 0$ and $n = 1$, giving the simpler relation:

$$\dot{\bar{\varepsilon}}^{cr} = A\tilde{q} \tag{0}$$

with its contribution to the overall framework controlled by a stiffness ratio, *s*. Values of A and *s* for each network were chosen such that the overall material behaviour was matched to data for PEEK melt at 380˚C, as outlined in the next section. Over the four networks, *s* summed to one, imparting Newtonian behaviour below 10 s^{-1}. Model parameters are provided in the Supplementary Information. Most models in the literature treat polymer melts as incompressible [30]. A Lagrangian approach employed in Abaqus/Standard®, in conjunction with a hybrid-element formulation, was chosen to capture this behaviour. Hybrid elements include additional nodes to facilitate the accurate calculation of pressure when there no change in volume. The use of

Abaqus/Explicit® with either a Lagrangian or Coupled Eulerian-Lagrangian approach was also explored, but both unfortunately led to non-physical changes in volume at high rates of strain.

2.1.1 *Calibration of PEEK melt model*

The material model was calibrated to match the experimental data for PEEK shear viscosity at 380°C [8], in a series of plane-strain, single-element tests. It can be seen in Fig. 3(a) that the computed shear viscosity at 1% shear strain amplitude, as a function of engineering shear strain rate, $\dot{\gamma}$, matches the data both in the Newtonian region, and in the shear-thinning region. In Fig. 3(b), a constant value of simulated shear viscosity is shown as a function of strain amplitude up to 300%, at a strain rate of 10 s^{-1} (within the Newtonian region), in line with its linear viscoelastic definition.

There is, to our knowledge, limited extensional data for PEEK in the literature [31]. However, during shear deformation of CF/PEEK, large positive and negative planar extensional strains occur in the small spaces between fibres. It is thus of interest to know if the material model captures this behaviour in at least a broadly reasonable manner. Planar extensional viscosity of the material model, η_E, was computed from the single-element extensional test illustrated in the inset in Fig. 3(a), using true strain, as is standard for viscoelastic fluids [32]. It can be seen from Fig. 3(a), for which all strain amplitudes are 1%, that in the Newtonian region, η_E is ~1672 Pa.s, while η is ~421 Pa.s, giving a Trouton ratio, T_R, of 3.97. The theoretical value of T_R for planar extension of a Newtonian fluid is four [33]. Thus, the extensional behaviour of our model is in line with theory for Newtonian fluids.

2.2 Two-fibre models

To explore the interplay between boundary-lubricated fibre friction and melt viscosity, two-dimensional, plane-strain, "two-fibre models" were constructed, consisting of two fibres immersed a in PEEK melt calibrated for 380°C, as illustrated in Fig. 4(a). The dimensions of the melt relative to the fibres correspond to a volume fraction of 62%. The carbon fibres were modelled as separate parts with 5 μm diameter, and transversely-isotropic, linear-elastic material properties. Quadratic triangular (CPE6H) and quadrilateral (CPE8H) hybrid elements were used for the melt, with a mesh seed size of 0.1 μm, except in the small spaces between fibres where it was 0.01 μm - 0.02 μm. Quasi-static analysis using the Abaqus/Standard® *VISCO keyword was employed, which accounts for time-dependency of the viscoelastic polymer melt flow in a quasi-static analysis. Periodic boundary conditions (PBCs) were applied via constraint equations to *North*, *South*, *East* and *West* node sets, in conjunction with the n_1, n_2, n_3, n_4 control nodes, Fig. 4, using an approach similar to [34]. PBCs enable representation of the overall CF/PEEK material, albeit with an artificially-regular fibre spacing and infinite extent. The lateral distance between fibres was set to 0.7 μm, and prevented from changing by constraining the *South* boundary. Since fibres are continuous in CF/PEEK, with some degree of tow twisting and entanglement, they have a high rotational stiffness. Hence, we prevented fibre rotation via constraint equations. A model variant with no polymer melt was also used.

Special consideration was given to how fibre-fibre interactions, fibre-melt interactions and melt flow could all be handled in the same Lagrangian model. Normally, for the fibres to come into frictional contact, the volume of the PEEK elements between fibres would have to drop to zero, leading to numerical problems. Our solution was to define a *fibre-fibre* contact interaction with a minimum fibre separation, via a contact clearance, d_{cl}, of 0.01 μm. Thus, fibre interaction occurred over regions of the fibre surfaces that came within 0.01 μm of each other. A second no-slip, no-separation, *fibre-melt* contact interaction was defined around the circumference of each fibre. Thus fibre-fibre and fibre-melt interactions, together with melt flow could all be captured in the one model, without numerical problems. The Coulomb friction model within Abaqus® was used with a simple rate-independent coefficient of friction, μ_f. Concerning the choice of friction coefficient, Roselman and Tabor [35] measured a dry coefficient of friction of crossing carbon fibres with an applied normal force between them, reporting values that ranged from 7 for low normal loads of 0.1 μN to 0.05 for higher normal loads of 1 mN and above. Servais et al. [19] reported a dry coefficient of friction for glass fibres of 0.29 that reduced to 0.08 when lubricated in a polypropylene melt at 200˚C. Since it is being proposed here that the fibres are boundary lubricated, lower values in the range of 0.05 – 0.1 were investigated. It was found that fixing $\mu_f = 0.05$ in all the results presented below, while varying the normal force, enabled matching of the experimental data in [8].

Concerning loading conditions, Fibre 2 was initially placed just out of numerical contact with Fibre 1 (i.e. d slightly larger than d_{cl}), and slightly to the left of vertical alignment with Fibre 1, as shown in Fig. 4(a). Two sequential loading steps were then applied. In Step 1, a normal force, F_{N}, was applied to the Fibre 2 reference node in the negative *y*-direction, pushing it into numerical contact with Fibre 1. The model now represents two fibres in frictional contact in the as-manufactured pre-preg material, with a normal force between them resulting from entanglement. In Step 2, F_{N} remained applied, and the Fibre 2 reference node was given a prescribed displacement in the positive *x*-direction at constant sliding velocity, v_s, with its vertical movement left free. This caused Fibre 2 to slide around Fibre 1, deforming the polymer melt with it. A global shear strain rate for the model (as opposed to local rates in individual melt elements) was then determined from:

$$\dot{\gamma}_{\mathrm{TwoFibre}} = \frac{v_s}{h_f} \tag{0}$$

where h_f is the vertical distance between the fibre centres at the instant when they are vertically aligned, as shown in Fig. 4(b). At the same instant, the reaction force on Fibre 1 in the *x*-direction, F_x, was recorded, and the "two-fibre model viscosity" was then determined as:

$$\eta_{\mathrm{TwoFibre}} = \frac{F_x / A_{\mathrm{TwoFibre}}}{\dot{\gamma}_{\mathrm{TwoFibre}}} \tag{0}$$

where A_{TwoFibre} is the cross-sectional area of the two-fibre model, given by the product of the model thickness, 1.0 μm, and width, 5.7 μm. For various values of F_{N}, a range of v_s values were simulated, corresponding to shear rates from 0.001 s^{-1} to 1000 s^{-1}. The spacing between fibres, d, was also recorded at the instant when the

fibres are vertically aligned to check for signs of "lift-off", i.e. increased fluid force at high shear rates causing the fibres to be pushed apart, potentially leading to mixed, or hydrodynamically-lubricated friction, rather than boundary-lubricated friction. A typical model consisted of ~27,000 elements, with simulation times ranging from 80 minutes at high strain rates to ~4 hours at low strain rates, on 48 2.5 GHz Intel Ivy Bridge cores of the Irish high-performance national supercomputer (ICHEC).

2.3 RVE Model

A two-dimensional, 40 μm × 40 μm, plane-strain, periodic RVE model, with 54 fibres, more representative of CF/PEEK pre-preg material, was developed, as shown in Fig. 5(a). The nearest-neighbour algorithm (NNA) of Vaughan and McCarthy [36] was used, which is designed to give a realistic positioning of fibres in a high-volume-fraction composite. With some adaptation of the algorithm parameters, a ~60% volume fraction was obtained with a random distribution of fibre diameters around a mean of 5.2 μm, representative of Suprem™ CF/PEEK pre-preg. The distribution contained fibre pairs located at opposite boundaries (see coloured, boundary-fibre sets in Fig. 5(a)), suitable for application of PBCs. The contact clearance between fibres was set to $d_{cl} = 0.01$ μm, as in the two-fibre model. The fibre-melt boundary contact was set to no-slip, no-separation. Quadratic, triangular (CPE6H), hybrid elements were used for all simulations. The mesh seed size was 0.08 μm, except between selected fibres, initially close together, that experience a large separation during the simulation, where it was 0.04 μm. This resulted in a model with ~250,000 elements. PBCs constrained the *East* fibres and melt nodes to have the same displacement as the *West* fibres and melt nodes. To simulate shear deformation with platens gripping *all* top and bottom fibres, all *South* fibres were fixed, and all *North* fibre displacements were set equal to the displacement of control node n_4, which was given a prescribed displacement, $u_x(n_4)$, at a constant rate. Fibre rotation was prevented via constraint equations. A global shear stress, τ_{RVE}, was determined using:

$$\tau_{\mathrm{RVE}} = \frac{F_x(n_4)}{A_{\mathrm{RVE}}} \tag{1}$$

where $F_x(n_4)$ is the reaction force at n_4, in the *x*-direction, and $A_{\mathrm{RVE}} = 40$ μm^2 is the product of the width (40 μm) and nominal thickness (1 μm) of the RVE. The global engineering shear strain, γ_{RVE}, was calculated as:

$$\gamma_{\mathrm{RVE}} = \frac{u_x(n_4)}{L} \tag{2}$$

where L is the height of the RVE (40 μm). The strain rate $\dot{\gamma}_{\mathrm{RVE}}$ is then the applied γ_{RVE} divided by the simulation time t. The "RVE model viscosity" was determined as:

$$\eta_{\mathrm{RVE}} = \frac{\tau_{\mathrm{RVE}}}{\dot{\gamma}_{\mathrm{RVE}}} \tag{3}$$

Models were run for a shear strain amplitude of 5% (corresponding to rheometry measurements in [8]), and strain rate, $\dot{\gamma}_{\mathrm{RVE}}$, varying from 0.001 s^{-1} to 1000 s^{-1}. Simulation times ranged from one hour for $\dot{\gamma}_{\mathrm{RVE}} = 1000\ s^{-1}$, to ~90 hours for $\dot{\gamma}_{\mathrm{RVE}} = 0.001\ s^{-1}$, on 96 2.5 GHz Intel Ivy Bridge cores.

The incorporation of fibre friction effects into the RVE model, via the method used in the two-fibre model, proved not to be feasible. Friction can only operate between fibres in numerical contact (i.e. fibres within the contact clearance, d_{cl}, of each other). However, the degree of fibre contact in the RVE greatly under-represents the degree of contact in the three-dimensional material. Furthermore, in the real material, twisting and entanglement of fibres in the axial direction, results in normal forces between fibres where they are in contact, which cannot be captured by a two-dimensional model. Because of these two issues, the full effect of friction between fibres is not captured just by enabling fibre-fibre friction in the model.

Our solution to this problem was to employ an artificial representation of the frictional resistance to RVE deformation via non-linear "SPRING1" elements [26], attached to the fibres, as illustrated in Fig. 5(a) and (b). This concept was informed by the work of Gutowski [37], who employed a non-linear spring representation of resistance to motion of curved arched fibres in a polymer melt during consolidation. In the present work, the springs were calibrated to produce virtually zero deflection for applied force below $\mu_f F_{\mathrm{N}}$, and constant resistance, $F_{\mathrm{s}} = \mu_f F_{\mathrm{N}}$, for applied force above this value. Each fibre was attached to two springs, one acting in the *x*-degree of freedom, the other acting in the *y*-degree of freedom, enabling frictional resistance in all directions. The only exception was edge fibre pairs that participated in PBCs, for which only one of the fibres was attached to springs – thus 44 of the 54 fibres were attached to springs as indicated by the spring symbols in Fig. 5(a). Prior to implementing this solution in the RVE model, two versions of the *Two-Fibre* model were created without polymer to test the concept. One had fibre-fibre friction turned on, while the other had friction turned off and a spring element attached to the reference node of Fibre 2 (Fig. 4(a)). As will be shown in the results section, the two models gave near identical results.

Finally, to see if fibre positions and diameters returned by the NNA have much influence on the results, we examined four additional random configurations at low and high strain rates (0.01 s^{-1} and 1000 s^{-1}).

3. Results

3.1 Two-fibre model results

3.1.1 *Effect of fibre-fibre friction and melt viscosity on viscosity of composite at low applied pressure*

In this section, two-fibre model results are shown for one value of F_{N} (0.2 μN), which was calibrated to represent experiments in [8] for which the pressure applied by the rheometer plates was low (104 kPa or below). This represents the normal force arising from twisting and entanglement, together with the additional normal force from applied pressure (which should be small at low pressure).

Examining Fig. 6(a), *with no polymer melt* in the model, the resistance force, F_x, while moving Fibre 2

over Fibre 1, is independent of shear rate and equal to $\mu_f F_N$ (0.05 × 0.2 μN = 0.01 μN). From equation (0), this behaviour corresponds to a viscosity that decreases with strain rate at a constant rate, i.e. power-law, shear-thinning behaviour, which is confirmed by the plot of η_{TwoFibre} in Fig. 6(b). Shown also in Fig. 6(b) is the experimental data at low pressure from [8], and data from Stanley and Mallon [4] and Groves et al. [5], for which the loading was close to simple shear. It can be seen that, with the chosen F_N value (and $\mu_f = 0.05$), the viscosity from this purely frictional process (i.e. with no polymer present), compares well with the experimental data for strain rates between 0.001 s^{-1} and 0.1 s^{-1}.

With both friction and polymer melt included, the values of F_x, Fig. 6(a), and η_{TwoFibre}, Fig. 6(b), are virtually identical to the friction-only case for strain rates up to about 0.01 s^{-1}. Thus, at low strain rates, although polymer melt is present, fibres are still in boundary-lubricated contact, and fibre friction is providing almost all the resistance to deformation, with little resistance coming from shearing of the fluid. As the strain rate increases, F_x, Fig. 6(a), increases. By 0.1 s^{-1} it has approximately doubled to 0.02 μN, and thereafter it settles into a near-linear rate of increase until ~100 s^{-1}, beyond which it starts to level off again. Correspondingly, η_{TwoFibre}, Fig. 6(b), exhibits approximately Newtonian behaviour between 0.1 s^{-1} and 100 s^{-1}, and becomes shear-thinning thereafter. At high strain rates, the resistance force, F_x, Fig. 6(a), is over three orders of magnitude higher than the frictional resistance, indicating resistance from shearing of the fluid is now dominant.

At high strain rates, it might be expected that increased pressure in the PEEK melt could result in fibres initially in contact being pushed apart, eliminating frictional interaction, and resulting in mixed and/or hydrodynamic lubrication with reduced resistance force, F_x. To examine this, the vertical contact force on Fibre 2 *from the melt*, $F_{y,FM}$, was obtained by summing the *y*-components of the *fibre-melt* contact forces, over the surface nodes of Fibre 2. This can be thought of as a "lift force" from the melt, attempting to lift Fibre 2 off Fibre 1. In addition, the vertical contact force on Fibre 2 *from Fibre 1*, $F_{y,FF}$, was obtained by summing the *y*-components of the *fibre-fibre* contact forces over the Fibre 2 surface nodes. The results are shown in Fig. 6(c). Below 100 s^{-1}, $F_{y,FM}$ is very small and negative, indicating a small adhesive force drawing the fibres closer together. These values do not appear on the figure due to the log scale used. At ~100 s^{-1}, $F_{y,FM}$ becomes positive and begins to rise to more substantial values, accompanied by a reduced value of $F_{y,FF}$. As a check on the validity of the results, it was confirmed that $F_{y,FM} + F_{y,FF} = F_N$ (0.2 μN) at all data points. The numerical contact distance also starts to rise at 100 s^{-1}, but only reaches ~0.00015 μm (0.15 nm) at 1000 s^{-1}. Shear-thinning of the melt is expected to limit further increases in lift force above 1000 s^{-1}. The simulated rise would be somewhat sensitive to details of the contact interaction but, by way of context, the surface roughness of a carbon fibre is in the range of 10-100 nm [38]. Thus, while lift-off is predicted to occur above ~100 s^{-1}, separation is expected to be small, and is unlikely to grow with increasing strain rate.

In Fig. 6(d), plots are shown of the total *horizontal* contact forces on Fibre 2, from the fibre-melt

interaction, $F_{x,FM}$, and from the fibre-fibre interaction, $F_{x,FF}$. As a check, it was verified that $F_{x,FM} + F_{x,FF} = F_x$ at all data points, i.e. the *x*-components of the contact forces from the two contact interactions add up to the total resistance to Fibre 2 motion. Here, we see clearly that frictional resistance dominates below 0.1 s^{-1}, while viscous resistance dominates thereafter. At 100 s^{-1}, the frictional resistance starts to reduce, due to partial lift-off. But by this strain rate, the viscous resistance is nearly three orders of magnitude larger than the frictional resistance, so the loss in frictional resistance is immaterial to the overall response. In conclusion, minor lift-off is predicted at strain rates above 100 s^{-1}, but should make negligible difference to the overall material response.

Overall, the two-fibre model shows that, as postulated in [8], the shear-thinning behaviour of CF/PEEK at low strain rates (see Fig. 1) can be replicated by including boundary-lubricated friction in the model. A boundary-lubricated friction coefficient of $\mu_f = 0.05$, together with a normal force of 0.2 μN between fibres in contact, provides a good match with experimental data at low applied pressure.

3.1.2 *Calibration of spring to represent friction in RVE model*

As outlined in Section 2.3, to represent friction in the RVE model, a non-linear spring was used to resist fibre motion. The spring was calibrated using the two-fibre model. The process is illustrated in Fig. 7(a), for one strain rate, 0.001 s^{-1}, with no polymer present. As can be seen, the spring parameters were tuned so that the spring force, F_s (green dashed line), rises quickly over a very small displacement (0.1 μm), and then plateaus at $F_s = 0.01$ μN, which is $\mu_f F_N$. In one two-fibre model, Fibre 2 was displaced by 5 μm around Fibre 1, with friction switched on, and the resisting force, "F_x with friction", was recorded continuously (blue line). In a second two-fibre model, the spring was attached to Fibre 2, and the simulation was repeated with friction switched off, giving "F_x with spring (no friction)" (red line). In both models, some "noise" occurs as Fibre 2 slides on Fibre 1, due to numerical chatter in the contact interaction. However, the average resisting force is the same in both models. In Fig. 7(b), results from the above two-fibre models, with polymer included, are shown for a large range of strain rates. The viscosity from the "with spring, no friction" model matches that of the "with friction, no spring" over all strain rates.

3.2 RVE model results

3.2.1 *Viscosity and shear stress versus shear rate*

RVE behaviour for $\dot{\gamma}_{RVE}$ from 0.001 s^{-1} to 1000 s^{-1} was examined. To assist the discussion, five particular strain rates are referenced below: $\dot{\gamma}_1 = 0.01$ s^{-1}, $\dot{\gamma}_2 = 0.5$ s^{-1}, $\dot{\gamma}_3 = 4$ s^{-1}, $\dot{\gamma}_4 = 100$ s^{-1}, and $\dot{\gamma}_5 = 1000$ s^{-1}.

Fig. 8(a) shows viscosity, η_{RVE}, versus shear rate, $\dot{\gamma}_{RVE}$, overlaid with data from experiments in which a low pressure was applied to the sample by the rheometer plates (52 kPa), [8], and data from Groves et al. [5]. *Without springs representing friction, it can be seen that the model is incapable of reproducing the power-law, shear-thinning behaviour observed experimentally at low strain rates*, instead predicting Newtonian behaviour. With friction included, the model is able to correctly predict the shear-thinning behaviour. The results match the

experimental data for strain rates up to about $\dot{\gamma}_2 = 0.5$ s^{-1}. Considering the complexity of the problem and the test procedure, and the simplifications in the model, the agreement is remarkably good. Results are significantly lower than data from experiments with a high applied pressure (313 kPa) [8] (Fig. 8(a)), as would be expected, since no external pressure was applied in the model. Fibre friction thus explains the yield-stress behaviour of CF/PEEK at low strain rates.

Above $\dot{\gamma}_3 = 4$ s^{-1}, the model exhibits viscous behaviour. The viscosity follows the shape of the PEEK curve, being approximately Newtonian from $\dot{\gamma}_3 = 4$ s^{-1} to $\dot{\gamma}_4 = 100$ s^{-1} and shear-thinning thereafter. It is however, an order of magnitude larger relative to PEEK, due to the high shear rate of the PEEK melt between fibres [15]. The error bars at $\dot{\gamma}_1 = 0.01$ s^{-1} and $\dot{\gamma}_5 = 1000$ s^{-1} show the variation ($\pm$ one standard deviation of the logarithm of η_{RVE}) over five nearest-neighbour algorithm configurations, confirming relative insensitivity to fibre arrangement.

The shear stress, τ_{RVE}, plotted in Fig. 8(b), exhibits trends in line with the above observations on viscosity. The variation in τ_{RVE} is relatively small up to $\dot{\gamma}_2 = 0.5$ s^{-1}, indicating shear-thinning behaviour. It then ramps up and is approximately linear with strain rate between $\dot{\gamma}_3 = 4$ s^{-1} and $\dot{\gamma}_4 = 100$ s^{-1}, indicating a Newtonian response. Above $\dot{\gamma}_4 = 100$ s^{-1}, it starts to level out, indicating a return to shear-thinning behaviour.

3.2.2 *RVE deformation, shear banding and friction contribution to material response*

Møller et al. [25], have shown that glassy materials that exhibit a yield stress, display strain localisation and shear banding under a shear load. To characterise the degree of shear banding, they plotted an experimentally determined "fraction of sheared material" versus strain rate. Shear banding is also observed in the RVE model. Analogous to Møller et al. [25], Fig. 9 shows the "fraction of fibres that move", f_{move}, versus $\dot{\gamma}_{\mathrm{RVE}}$ (pink triangles). The "fibres that move" are a subset of the 44 fibres with springs attached representing friction (Fig. 5(a)), for which the force in the *x*-spring reached $F_s = 0.01$ μN, see Fig. 7(a). In other words, they are the fibres which have overcome frictional resistance and started to "slide". They are shown cross-hatched in the insets, which illustrate the deformation for $\dot{\gamma}_1$, $\dot{\gamma}_2$ and $\dot{\gamma}_5$. The insets show clearly that shear banding occurs at low strain rates. For example, at $\dot{\gamma}_1 = 0.01$ s^{-1} only a few fibres at the top move, while the material below this layer does not flow, and instead acts like a solid. In contrast at high strain rates, e.g. $\dot{\gamma}_5 = 1000$ s^{-1}, all fibres move, and the material flows everywhere. Quantitatively, for strain rates below $\dot{\gamma}_1 = 0.01$ s^{-1}, $f_{move} \leq 0.25$. An S-shaped transition then occurs to $f_{move} = 1$ at $\dot{\gamma}_4 = 100$ s^{-1} and above.

The stress, τ_{RVE}, arises from resistance provided by fibre friction and polymer deformation. The portion caused by fibre frictional resistance, τ_{fric}, was calculated in a global sense as:

$$\tau_{fric} = \frac{\sum f_{x_i}}{A_{\mathrm{RVE}}} \tag{3}$$

where f_{x_i} is the force in the *x*-spring connected to fibre *i*. This stress is plotted in Fig. 8(b) (grey circles). Additionally, the ratio $R_f = \tau_{fric}/\tau_{\mathrm{RVE}}$, a measure of the extent to which the material response is dominated by friction, is plotted in Fig. 9 (blue squares). It is seen that at strain rates up to $\dot{\gamma}_2 = 0.5$ s^{-1}, $R_f \cong 1$, indicating that virtually all the resistance is supplied by fibre friction, with the melt contribution apparently being negligible (see also Fig. 8(b), where $\tau_{\mathrm{RVE}} \cong \tau_{fric}$). In this regime, the material is behaving as a "yield-stress" material, and τ_{RVE} is the yield stress in shear, τ_y. A point to note though is that in this strain-rate regime, τ_y increases with strain rate – see Fig. 8(b). It does so because, as the strain rate increases, the melt becomes progressively stiffer and hence, more capable of transferring load between fibres. Consequently, more fibres are forced into motion - see the Fig. 9 insets at $\dot{\gamma}_1$ and $\dot{\gamma}_2$ - leading to *increased frictional resistance*. For example, at $\dot{\gamma}_2 = 0.5$ s^{-1}, we see from Fig. 9 that $f_{move} \cong 0.63$, compared to just 0.25 at $\dot{\gamma}_1 = 0.01$ s^{-1}. Thus, even though $R_f \cong 1$ up to $\dot{\gamma}_2 = 0.5$ s^{-1}, and the melt appears to contribute negligibly to the externally measured stress, it is in fact significantly influencing the response by enabling more and more fibres to engage in frictional resistance as the strain rate increases. In essence, *due to shear-banding*, the material has a *strain-rate dependent yield stress* in shear, τ_y, in the yield-stress, strain-rate regime.

From Fig. 8(b), at $\dot{\gamma}_2 = 0.5$ s^{-1}, τ_{fric} starts to level off. Although not clear on the log scale shown it does not fully level off until $\dot{\gamma}_4 = 100$ s^{-1}. Fig. 9 shows *why* τ_{fric} levels off in this way – it is because the rate of increase in f_{move} with strain rate starts to decrease. In particular, by 2 s^{-1}, 42 of the 44 fibres with springs (i.e. 95.5%) are in motion. Relatively large increases in strain rate are required to get the final two fibres to move. These are Fibre 5 which moves at 10 s^{-1}, and Fibre 16 which moves at $\dot{\gamma}_4 = 100$ s^{-1} (see inset at $\dot{\gamma}_5 = 1000$ s^{-1} for fibre numbering). These two fibres are in the bottom row of non-fixed fibres (just above the fixed row at the bottom), and of the fibres in that row, they are the ones most wedged in by fibres to their right. Shear banding is thus fully eliminated at $\dot{\gamma}_4 = 100$ s^{-1}. Note again that no external pressure is applied in the model, so these findings on shear banding are most relevant to the low pressure experimental case (52 kPa) [8].

Referring again to τ_{RVE} in Fig. 8(b) and R_f in Fig. 9, once the frictional resistance levels off, the material transitions to viscous behaviour. By $\dot{\gamma}_4 = 100$ s^{-1} the transition is essentially complete, $R_f \cong 0.025$, and the frictional contribution to resistance is negligible.

3.2.3 *Load transfer from melt to fibre in the RVE*

In this section, the mechanisms by which load is transferred within the melt to move fibres are examined. Borrowing from the field of aerodynamics, we calculate the *pressure drag* and *skin-friction drag* on each fibre. Pressure drag, resulting from a pressure gradient in the surrounding melt, is found by considering the *x*-components of the contact *normal* forces, N_x, on the fibre edge nodes coming from the fibre-melt contact interaction (CFN1 in Abaqus®). With no external pressure applied, pressure in the melt arises from deviatoric melt stress, sometimes called "extra stress". Skin-friction drag is obtained from the *x*-components of the contact

shear forces, S_x (CFS1 in Abaqus®). Fig. 10(a) is focused on Fibre 39 which is at about the vertical midpoint of the RVE (see Fig. 9), and shows σ_{rr}, the radial stress in the melt with reference to a local coordinate system at the fibre centre, and $\tau_{r\theta}$ in the same coordinate system. The plot of σ_{rr} is virtually the inverse of a plot of pressure (since σ_{rr}, $\sigma_{\theta\theta}$ and σ_{zz} are all similar in value), but its effect is more intuitive to visualise since positive σ_{rr} (negative pressure) indicates an area where the melt is in tension and *pulling* on the fibre. Shown also are vectors representing N_x and S_x, together with the resultant pressure drag force $D_p = \sum N_x$, and resultant skin-friction drag force $D_s = \sum S_x$. Fig. 10(b) gives the same information for Fibre 16 (the last fibre to move). The strain rate in Fig. 10 is $\dot{\gamma}_4 =$ 100 s^{-1}, the lowest strain rate at which all fibres are in motion. Note that the resultant force in the *y*-direction and resultant couple are small and not shown.

Examining Fig. 10, it can be seen that high stresses (both tensile and compressive) are induced in the melt in the small spaces between fibres, more than an order of magnitude higher than the global stress, τ_{RVE}, which is $\cong 5 \cdot 10^5$ Pa at $\dot{\gamma}_4 =$ 100 s^{-1} (see Fig. 8(b)). As an aside, the model assumes the melt adhesion to the fibres can sustain high tensile stresses without "failing", but clearly large tensile stresses at the fibre-melt interface could lead to the formation of new (i.e. not pre-existing) voids in the material.

Regarding Fibre 39, Fig. 10(a), the most prominent aspect of the pressure drag force (N_x) distribution is the region of large positive values at the space between Fibres 39 and 40. Essentially, Fibre 40, which is moving to the right, is "dragging" Fibre 39 after it, with the load being transferred between the two fibres via large tensile stresses (i.e. negative pressure) in the melt. The very small space between these two fibres leads to a large amplification of the stresses in the melt, and drag forces on the fibre. Acting against this movement are N_x forces arising from compression of the melt between Fibres 39 and 27, and from tension in the melt between Fibres 39 and 26. Overall though, the net pressure drag force $D_p =$ 0.53 μN is positive (i.e. to the right). The shear stress values, $\tau_{r\theta}$, are considerably smaller than the σ_{rr} values, and consequently the skin-friction drag forces, S_x, are also smaller in magnitude than the N_x forces. The net skin-friction drag force, $D_s =$ -0.52 μN, is negative (to the left) primarily due to the region at the bottom of the fibre, where shear stresses opposing Fibre 39 motion arise in the melt, as Fibre 39 slides across Fibres 26 and 27. The total drag force on the fibre, $D = D_p + D_s =$ (0.53 - 0.52) μN, i.e. $D =$ 0.01 μN, as it has to be when the fibre is moving, since the spring representing friction provides a constant resisting force $F_s =$ 0.01 μN (see Fig. 7(a)).

Turning to Fibre 16 in Fig. 10(b), a region of positive N_x values exists between Fibres 16 and 7, due to tensile stresses in the melt as Fibre 7 pulls Fibre 16 in its direction of motion. This is resisted by N_x forces arising from compression of the melt between Fibres 16 and 29, and tension in the melt between Fibres 16 and 28. Overall, the net pressure drag $D_p =$ -0.75 μN is negative. This time, it is the skin-friction drag forces which tip the scale in favour of movement to the right, and $D_s =$ 0.76 μN is positive, primarily due to the movement

of the fibre above (Fibre 38) which generates shear stresses in the melt that drag Fibre 16 along. Once again, the total drag force $D =$ 0.01 μN since Fibre 16 is in motion.

The variation of drag forces on these two fibres with strain rate is shown in Fig. 11. At low strain rates the drag forces are small, and the total drag force *D* varies with strain rate and is less than the value required to overcome fibre "friction" (i.e. $D < F_s$). Fibre 39 starts to move at $\dot{\gamma}_2 =$ 0.5 s^{-1}, and thereafter *D* is constant ($D = F_s$). For Fibre 16, *the last fibre to move*, the total drag force *D* does not reach F_s until $\dot{\gamma}_4 =$ 100 s^{-1}. However, from $\dot{\gamma}_3 =$ 4 s^{-1} onwards the difference between *D* and F_s is extremely small, and given that F_s is an approximate number representing fibre sliding, one can say that shear-banding is predicted to be essentially eliminated by $\dot{\gamma}_3 =$ ~4 s^{-1}.

Once fibres move, the pressure and skin-friction drag forces, D_p and D_s, are always of opposite sign. Some fibres, coloured red in the Fig. 11 inset, are moved primarily by skin-friction drag (i.e. D_s is positive), and others, coloured green, are primarily pushed or pulled by pressure drag forces (i.e. D_p is positive). Which mechanism dominates for a given fibre, depends on the positions of the surrounding fibres.

An interesting correlation between melt pressure and shear banding arises as follows. The average value of the *required pressure differential across a fibre* to overcome fibre friction resistance, F_s, can be very approximately calculated as the frictional sliding force divided by the projected area of an average fibre, i.e.

$$\Delta P_{req} = \frac{F_s}{d_{avg} t} \tag{3}$$

With an average fibre diameter, $d_{avg} =$ 5.2 μm, an RVE nominal thickness of 1 μm and $F_s =$ 0.01 μN, this gives $\Delta P_{req} =$ 1.92 kPa. Such a calculation of course ignores the complex shear and normal stress interactions noted above. Fig. 12 shows the mean of the logarithm of positive (red) and negative (blue) pressure in the melt, with bars indicating ± three standard deviations, within which, 99.7 % of pressure data lies. These are plotted as a function of strain rate, together with a green line representing ΔP_{req}. At low strain rate, $\dot{\gamma}_1 =$ 0.01 s^{-1}, the inset illustrates that melt pressure in the vast majority of the RVE is negative. Over this large region of negative pressure, the mean pressure is ~2 kPa, and the range of pressures, $P^-_{range} = \left|P^-_{+3\sigma}\right| - \left|P^-_{-3\sigma}\right|$, is relatively small at ~8 kPa. A large positive pressure range $P^+_{range} = P^+_{+3\sigma} - P^+_{-3\sigma}$, of ~200 kPa exists, but is localised only in a small area of the melt. Thus, in much of the melt, $|P| < \Delta P_{req}$, and the small range implies that the likelihood of the pressure changing by ΔP_{req} across a single fibre is low. Thus, we expect that not many fibres will move, which concurs with the figure of $f_{move} \cong 0.25$ in Fig. 9.

At $\dot{\gamma}_2 =$ 0.5 s^{-1}, P^-_{range} has increased ten-fold to ~80 kPa, while P^+_{range} is still ~200 kPa, but exists over a larger area of the melt. This means the likelihood of the pressure differential across any given fibre exceeding ΔP_{req} is higher. This is borne out by the figure of $f_{move} \cong 0.63$ from Fig. 9. However, there are still large areas of

the melt where $|P| < \Delta P_{req}$. In such areas, developing a pressure differential of ΔP_{req} across a fibre is unlikely, so some fibres do not move. At $\dot{\gamma}_3 =$ 4 s^{-1}, the positive and negative pressure ranges are both ~700 kPa, which is much higher than at $\dot{\gamma}_2 =$ 0.5 s^{-1}. In addition, $|P| > \Delta P_{req}$ in the vast majority of the melt. The much larger range of pressures will in general lead to much larger pressure gradients. Furthermore, with most of the melt now at a pressure above ΔP_{req} the likelihood of a pressure gradient of ΔP_{req} existing across all fibres is very high. This concurs with Fig. 9 which shows a value of $f_{move} = 0.955$. Finally, at $\dot{\gamma}_4 =$ 100 s^{-1}, $|P| > \Delta P_{req}$ everywhere and the pressure range is even larger. Now pressure gradients are large everywhere, so all fibres should move, which agrees with Fig. 9 where $f_{move} = 1$. In summary, a higher strain rate leads to higher pressures, *and* a larger range of pressures, resulting in larger pressure gradients, and hence increased likelihood that all fibres will move. An approximate, empirical criterion for the elimination of shear-banding appears to be $|P|_{\min} > \Delta P_{req}$, where $|P|_{\min}$ is the lowest (i.e. -3σ value) pressure in the melt.

3.2.4 *Homogenised viscosity, $\eta_{y_{\max}}$, in yield stress regime*

The occurrence of shear banding in a shearing test is undesirable, since the full material thickness is not tested. As seen above, shear banding can be eliminated if melt pressure is sufficiently high for the approximate criterion $|P|_{\min} > \Delta P_{req}$ to be met. This criterion is met at strain rates above (approximately) $\dot{\gamma}_3 =$ 4 s^{-1}. This melt pressure results from high, deviatoric stresses (extensional and compressive) in the melt, and is expected to be relatively constant across the material and independent of sample width. Another way to increase the pressure in the melt is to apply external pressure on the sample. According to Fig. 12, the average pressure in the melt at $\dot{\gamma}_3 =$ 4 s^{-1}, where the criterion $|P|_{\min} > \Delta P_{req}$ is approximately met, is ~30 kPa. This provides a very approximate prediction for the external pressure required to eliminate shear banding at low strain rates. However, externally applied pressure takes on high values at the sample centre, that decrease to low values at the sample edge [3]. Consequently, elevated pressure, higher than 30 kPa, may be necessary to ensure the pressure gradient exceeds ΔP_{req} all the way to the sample edge. The experimental tests in [8] showed that CF/PEEK transverse-shear viscosity was about one order of magnitude higher at pressures above 156 kPa than it was at pressures below 104 kPa. We postulated that the change in viscosity between 104 and 156 kPa was due to the behaviour transitioning from a shear-banded to a non-shear-banded response. Thus, the ~30 kPa prediction is of a similar order of magnitude to, but, as expected, slightly lower than the external pressure where the aforementioned change in viscosity occurred, lending credence to the postulate that the viscosity change is due to the elimination of shear banding.

We conclude from this that the experiments in [8] at applied pressures below ~104 kPa [8] are not valid material tests, since shear banding is occurring. Only tests for which the applied pressure was sufficient to eliminate shear banding, are valid. In such tests, once the yield stress in shear is exceeded, all fibres move, and the corresponding viscosity can be considered the "homogenised material" viscosity. We call this $\eta_{y_{\max}}$ - the viscosity at a given strain rate, if sufficient pressure is applied such that all fibres move. It is not possible to

study external pressure in our RVE model, due to the PBCs applied. A model of the full width and thickness of the tape would be required, which was not feasible with available computing resources. However, $\eta_{y_{\max}}$ can be deduced from the model as follows. As seen in Fig. 9, $f_{move} = 0.955$ at $\dot{\gamma}_3 = 4$ s^{-1}, meaning shear banding is effectively eliminated. The corresponding τ_{RVE} is the stress the RVE has to be capable of sustaining, in order to overcome the frictional resistance of all fibres. It is the yield stress when all fibres move, or the yield stress corresponding to $\eta_{y_{\max}}$. We call this $\tau_{y_{\max}}$, and it is seen that $\tau_{y_{\max}} = \tau_y\left(\dot{\gamma}_3\right)$. It can be noted from Fig. 8(b) and Fig. 9 that the melt is contributing a large part of τ_{RVE} at $\dot{\gamma}_3 = 4$ s^{-1}. It *has* to be capable of contributing this level of resistance in order to transfer the applied load all the way to the last fibre to move. The high deviatoric extensional and compressive stresses in the melt at this strain rate result in the high melt pressure, and enable the transfer of more load. At lower strain rates, it would *still* have to be capable of contributing this level of stress in order to move all fibres. To make this possible, external pressure would have to be applied, to bring the melt pressure, comprising extensional and compressive stress components, up to the value needed to transfer the load to the last fibre. Thus, to move all fibres at strain rates below $\dot{\gamma}_3 = 4$ s^{-1}, a shear stress of $\tau_{y_{\max}}$ must be sustained by the material. This observation is illustrated by the dashed blue line on Fig. 8(b). Corresponding to this shear stress, which is constant with strain rate, is a linearly varying viscosity, which is $\eta_{y_{\max}}$, as shown by the dashed blue line in Fig. 8(a). We note that the $\eta_{y_{\max}}$ line is in remarkably good agreement with the experimental data in [8], for which the external pressure was high (313 kPa).

3.3 Relevance for Automated Tape Placement processing

One of the main purposes of this work was to characterise CF/PEEK behaviour to enable development of better ATP processing models. ATP processing involves squeeze flow of the tape, during which the transverse shear deformation studied here takes place. According to Lipscomb and Denn [39], continuity of velocity during squeeze flow requires that the yield stress be exceeded everywhere so that all the material flows. Hence, for ATP processing of CF/PEEK, *all* fibres in the material must displace. The strain rate varies widely in different regions of the tape, but for regions where the strain rate is below ~4 s^{-1} the material is responding as a yield-stress material, and since all fibres are moving, the relevant viscosity is $\eta_{y_{\max}}$. Thus, the experimental data [8], for which the external pressure was 313 kPa, is the data to use in ATP models (see Fig. 8(a)). At strain rates above ~4 s^{-1}, we predict viscous material behaviour, with a viscosity roughly an order of magnitude higher than PEEK (again see Fig. 8(a)). We also predict shear thinning above ~100 s^{-1}. It should be noted that experimental data at such high rates does not exist, and is probably not possible to generate with existing rotational rheometers. Nonetheless, strain rates far above this are predicted to occur during ATP processing (up to 2500 s^{-1} according to [27]), so it is important to have an idea of the material behaviour at such rates for modelling the ATP process.

Squeeze flow needs to occur in ATP processing to enable compression of voids, flattening of surface roughness to facilitate intimate contact between the tape and underlying laminate, and expansion in the width

direction to fill gaps between parallel tape segments. Thus, the yield stress in shear, $\tau_{y_{\max}}$, must be overcome at all points in the material [39], which means there is a global yield stress in compression, σ_y, which must be overcome by the ATP roller pressure for successful processing to occur. The value of σ_y is most likely influenced by pre-preg tape width. The pressure under the roller is contingent on the type of roller employed, with values of ~180 kPa being typical for a silicone roller [40], while much higher values are predicated for metal rollers [41]. Given compressive yield occurred in our tests in [8] at ~225 kPa, it is possible that consolidation would not occur under silicone rollers, if the applied pressure was too low.

4. Conclusions

The models presented above have demonstrated that the experimental behaviour of CF/PEEK seen in [8], and in previous work, can be explained by considering it to be a yield-stress material at low strain rates, transitioning between 0.1 – 100 s^{-1}, to a viscous material at high strain rates. Fibre friction is the dominant process in the yield-stress regime, but is relatively unimportant in the viscous regime. In the viscous regime, the viscosity is predicted to be that of PEEK, amplified by approximately one order of magnitude, due to the requirement for the melt to undergo high strain rates in the small inter-fibre spaces [15]. The results have also shown that shear-banding will occur below strain rates of about 4-5 s^{-1} if insufficient pressure is applied to the rheological sample. A pressure of at least 150 kPa is recommended for rheological tests at such strain rates, although it is likely this value would vary somewhat with specimen geometry. For continuum models of ATP processing, it is noted that Newtonian models of CF/PEEK are grossly inaccurate. A strain-rate dependent viscosity is imperative for accurate modelling, and for regions of the tape where the strain rate is below about 4 s^{-1} only the data from the tests in [8] under high applied pressure should be used.

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Figure Captions

Fig. 1

(a) CF/PEEK transverse shear viscosity from [8] at 5% strain amplitude, 380˚C, and both 52 kPa and 313 kPa applied pressure, together with data from previous tests in the literature, denoted A: Stanley and Mallon [4], B: Groves et al. [5], C: McGuinness and Ó Brádaigh [6] and D: Shuler and Advani [7].

Fig. 2

Transverse shear viscosity versus shear strain magnitude, for APC-2 CF/PEEK coupons, deformed in shear at a strain rate of 0.01 s^{-1}. Insets: schematic of specimen after shear strain of ~50%, together with image of specimen edge (adapted from Stanley and Mallon [4]).

Fig. 3

Single element PEEK response, (a) transverse and extensional viscosity versus strain rate at a strain amplitude of 1%, with inset showing two deformation modes, together with experimental data from [8] at 380˚C, (b) shear viscosity versus engineering strain, and extensional viscosity versus true strain, both at strain rate of 10 s^{-1}.

Fig. 4

Two-fibre model, (a) initial configuration with boundary conditions, fibre reference nodes, lateral spacing d_l =0.7 μm, fibre diameter d_f =5 μm, initial vertical spacing, *d*, slightly larger than contact clearance d_{cl} = 0.1 μm, and load steps, (b) instant when fibres are vertically aligned.

Fig. 5

(a) 40 μm × 40 μm RVE, showing nodesets for boundary conditions for simple shear test simulation, *North*, *South*, *East* and *West* fibre sets, fibres which have springs attached to represent friction, (b) schematic of non-linear spring used to represent fibre-fibre friction.

Fig. 6

Two-fibre model results, F_N = 0.2 μN, μ_f = 0.05, (a) resisting force, F_x, with/without polymer, (b) Transverse shear viscosity from model and experiments in [8] measured at 52 kPa applied pressure, and from the literature, (c) vertical contact forces on Fibre 2 from melt, $F_{y,FM}$ and from Fibre 1, $F_{y,FF}$, together with numerical contact distance between fibres, and (d) horizontal contact forces on Fibre 2 from melt, $F_{x,FM}$ and from Fibre 1, $F_{x,FF}$.

Fig. 7

Results from two-fibre models, one with friction enabled, and one with friction disabled but with a spring

attached to Fibre 2, (a) resisting force, F_x, with no polymer, for a 5 μm displacement around Fibre 1, at one strain rate, $\dot{\gamma} = 0.001$ s^{-1}, (b) viscosity versus strain rate, with polymer.

Fig. 8

Behaviour of RVE with/without springs, (a) η_{RVE} versus strain rate at strain amplitude of 5%, compared to experimental data from [8], at applied pressures of 52 kPa and 313 kPa, and simulated results for neat PEEK. Error bars at strain rates of 0.01 s^{-1} and 10^3 s^{-1} are for one standard deviation of the logarithm of η_{RVE}, for five different RVE configurations. Also shown is calculated $\eta_{y_{\max}}$, the viscosity in the yield stress regime when all fibres move, (b) τ_{RVE} versus strain rate at strain amplitude of 5%. Also shown is the calculated $\tau_{y_{\max}}$, the yield stress when all fibres move.

Fig. 9

Fraction of fibres in RVE that move, and fraction of external shear stress that is resisted by friction, both plotted versus strain rate at a strain amplitude of 5%. Insets show RVE deformation at strain amplitude of 5%, at $\dot{\gamma}_1$, $\dot{\gamma}_2$ and $\dot{\gamma}_5$, with cross-hatching to indicate fibres that have moved, and Fibres 5, 16 and 39 highlighted.

Fig. 10

Contours of σ_{rr} and $\tau_{r\theta}$ in the melt (coordinated system origin at presented fibre centre), *x*-components of contact-normal force, N_x, providing pressure-drag, and *x*-components of contact-shear force, S_x, providing skin-friction drag, for (a) Fibre 39, and (b) Fibre 16, both at strain rate of 100 s^{-1} and strain amplitude of 5%. Red arrows indicate total pressure-drag, $D_p = \sum N_x$, and skin-friction-drag, $D_s = \sum S_x$, in *x*-direction.

Fig. 11

Drag force versus strain rate in positive and negative *x*-directions, showing pressure-drag, friction-drag and total-drag force for Fibres 39 and 16 of the RVE at 5% strain amplitude. Also shown is F_s, the spring force that the total drag force must exceed for fibre displacement to occur. Inset shows the drag force type that moves each fibre at 100 s^{-1}. Green indicates pressure drag, red indicates skin-friction drag. Note: Fibre 16 is the last to move.

Fig. 12

Mean of the logarithm of melt pressure, positive (red) and negative (blue), with bars indicating $\pm$ three standard deviations, versus strain rate at 5% strain amplitude, with insets illustrating sign of pressure. Shown also is line illustrating ΔP_{req}, an approximate estimate of the pressure differential across a fibre to move it against friction.