Prediction of the peak and steady-state ply-ply friction response for UD C/PAEK tapes

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\textbf{Abstract}

Excessive ply-ply friction can lead to process-induced defects during the hot press forming of thermoplastic composite laminates. Process simulations can be used to enable first-time-right defect-free manufacturing. However, a good understanding of the ply-ply friction is required to improve the constitutive models to allow simulation of more complex parts. We performed friction experiments on UD C/PAEK tapes, showing a typical response with a peak followed by a steady-state friction. Micrographs of tested specimens were analyzed to evaluate the matrix interlayer thickness distribution in the ply-ply interface, which we combined with the matrix viscosity to predict the peak friction. Generated fiber distributions, mimicking the ply-ply interface, were successfully used to obtain the matrix interlayer as well. The steady-state friction was accurately described by including a critical shear stress to represent wall slip, substantiating the concept of wall slip as the dominant underlying mechanism for the start-up friction response.

\textit{Keywords:} A. Polymer-matrix composites (PMCs); A. Thermoplastic resin; B. Rheological properties; E. Forming

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1. Introduction

Hot press forming is a rapid, automated, and cost-effective technology to manufacture continuous fiber-reinforced thermoplastic composite (TPC) parts. The process is already successfully applied for clips and brackets in the Boeing B787 and the Airbus A350 [1] and its application for the manufacture of parts in the primary structure is foreseen [2]. Despite its maturity, it can be challenging to set up a proper production line that manufactures defect-free parts. One of the issues to be addressed is forming-induced wrinkles, which result in weak spots and, consequently, rejection of parts [3, 4].

Process simulation software for hot press forming can be used to minimize the occurrence of wrinkles beforehand through virtual design iterations. Obviously, accurate predictions on defect generation are then required, which is challenging for complex geometries [2]. This situation is especially true for unidirectional (UD) reinforced materials, as forming experiments have shown that UD parts suffer from distortions not seen with woven fabrics [3, 5], of which some cannot be sufficiently predicted.

Predictive forming simulations are based on a careful mathematical description of the deformation mechanisms that are active during forming, most notably bending, intra-ply shear, and ply-ply and tool-ply slip [4]. Though all of these mechanisms are relevant [6], this study focuses on ply-ply friction, or the resistance against movement of adjacent plies. Ply-ply friction plays an important role in the formation of defects as it leads to compressive stresses in an inner radius [7] and it hinders intra-ply shear in parts with double curvature [8–10].

Ply-ply friction characterization experiments, in which often a single ply is pulled out of a stack of plies in the molten state, typically reveal a complex transient behavior with a peak or overshoot, $\tau_p$, followed by a steady-state or long-time shear stress, $\tau_\infty$, as schematically illustrated in Fig. 1. The friction response depends on temperature, normal pressure and, in particular, rate [11, 12]. Several studies included modeling of the friction behavior. In case of woven fabric reinforcements, the steady-state response can be described with
the Striebeck curve in the hydrodynamic lubrication regime [10, 13–16]. This approach, however, was found to be inapplicable to UD material [4, 9]. Hence, Sachs [4] proposed a model based on dry and viscous friction, including a critical shear stress for wall slip, to describe the steady-state tool-ply friction response of UD C/PEEK. Although the predicted response was of the same order as experimental observations, the prediction lacked accuracy as the rate-dependency was not well captured. Other modeling efforts include often a Coulomb friction or yield stress combined with a viscous law to fit the experimental data [7, 11, 12, 17].

The full transient (start-up) friction response was not treated in the models discussed above, which all focus on the steady-state regime. In practice, however, ply-ply slip occurs over only a few millimeters [3, 4, 17]. Hence, for most forming problems, the start-up response is actually more relevant than the steady-state regime. Only a few studies dealt with modeling of the full transient response. Fetfatsidis et al. [15] and Vanclooster [8] mimicked the response by using an exponential and a linear decay, respectively, from peak towards steady-state values, neglecting stress growth at start-up. Chen et al. [18] included stress growth with a model that originated from crack initiation simulations, but the transition from peak to steady-state shear stress is distorted as pull-out friction data were used.

Figure 1: Typical transient (start-up) ply-ply friction response of shear stress versus time.
A fundamental understanding of the mechanism responsible for the transient ply-ply friction response is, to the best of our knowledge, lacking. Recently, we evaluated different hypotheses for the transition from peak to long-time shear stress and suggested a slip relaxation effect, gradually giving rise to wall slip, as most probable [19]. This research aims to strengthen the concept of wall slip as the key underlying mechanism for ply-ply friction through modeling of the friction characteristics, i.e. the peak and long-time shear stress, based on the matrix viscosity and the ply morphology for UD C/PEEK and C/LM-PAEK. An improved understanding of ply-ply friction will help to develop a more accurate constitutive description for simulation software. In the end, the increased predictive quality will help to enable first-time-right defect-free manufacturing of UD fiber-reinforced thermoplastics to push the development of lightweight structures even further.

2. Experimental work

The materials considered in this research will be presented, followed by a brief description of the rheometry experiments performed on the neat matrix materials, the microscopy work on the ply-ply interfaces, and, lastly, the experimental methods used to measure ply-ply friction.

2.1. Materials

Two pre-impregnated composite material systems were considered: one with a PEEK and one with a LM-PAEK matrix, both reinforced with approximately 59% unidirectional carbon fibers per volume. The materials are known as Toray Cetex TC1200 (C/PEEK) and TC1225 (C/LM-PAEK), respectively. For C/PEEK, the melting temperature is 343°C and the processing temperature ranges from 370-400°C, whereas for C/LM-PAEK the melting temperature is 305°C and the processing temperature ranges from 340-385°C, as stated by the manufacturer Toray Advanced Composites [20].
2.2. Rheometry

The neat matrix materials, Victrex PEEK 150P and Victrex LM-PAEK AE250P, were subjected to plate-plate rheometry to measure their viscosities. The polymers were characterized in an Anton Paar Physica MCR501 rheometer equipped with a parallel-plate geometry. Frequency sweeps were performed in nitrogen atmosphere at 380 and 390 °C for PEEK specimens and at 365 °C for LM-PAEK specimens. A shear strain of 1% was used, which was well within the linear viscoelastic regime based on oscillatory amplitude sweeps. The modulus of the complex viscosity versus frequency data was used to obtain steady shear viscosity \( \eta \) versus shear rate \( \dot{\gamma} \) curves by applying the Cox-Merz rule, of which the validity was checked at low rates through continuous rotational experiments. The obtained viscosity curves were then fitted with the Cross model [21],

\[
\eta(\dot{\gamma}) = \frac{\eta_0}{1 + \left(\frac{\dot{\gamma}}{\tau_*}\right)^{1-n}}
\]

with the zero shear viscosity \( \eta_0 \), the critical shear stress \( \tau_* \), and the power law index \( n \) as fitting parameters.

2.3. Microscopy

The matrix interlayer thickness between adjacent plies will be nonuniform over the width of the specimen due to the fibers at the interface, which will affect the friction response. The exact morphology of the ply-ply interface is, therefore, required. Hence, tested ply-ply specimens were cut at three locations in the contact area perpendicular to the sliding direction using a water-cooled diamond saw. Nine cross-sections were taken from two C/PEEK specimens and one C/LM-PAEK specimen, which were embedded and polished. A Keyence VHX 7000 digital microscope was used to obtain 8 and 9 micrographs of the C/PEEK and C/LM-PAEK cross-sections, respectively. These micrographs were later used to analyze the upper and lower ply-ply interfaces.
2.4. Ply-ply friction tests

2.4.1. Test setup

Ply-ply friction measurements were conducted on a benchmarked friction tester [22], schematically shown in Fig. 2 and described in more detail in e.g. [4, 23, 24]. The pre-preg materials, supplied on 12-inch rolls, were first cut and stacked with all fibers aligned with the longitudinal or sliding direction, according to Fig. 2. The two outer plies, measuring 120-mm long and 50-mm wide, were clamped at the bottom of the test setup, while the central ply (250 × 50 mm²) was fixed in the upper clamp of a universal testing machine, in which the friction tester was positioned. Alignment of the specimen was checked using a laser line leveller. The temperature and normal pressure in the contact area were controlled through heated pressure platens, with applied normal force $F_n$ and area $A$ of 50 × 50 mm². The platens were shielded from the polymer melt using metal foils. Rate-controlled displacement of the crosshead induces a shearing action in the ply-ply interfaces, resulting in a pulling force, $F_{\text{pull}}$. The shear stress in the contact area per slip interface, $\tau$, can then be computed as follows:

$$\tau = \frac{F_{\text{pull}}}{2A}$$  \hspace{1cm} (2)

An additional overlap of 15 mm was used to ensure a constant contact area during the test, for which heating elements below the pressure plates provided a proper material temperature at the inlet. The pulling force was zeroed at the end of each run by recording the force while the specimen was freely hanging in the upper clamp.

2.4.2. Experimental procedure and conditions

The friction test procedure we followed is depicted in Fig. 3. After mounting a specimen, a normal pressure was applied and a waiting time of 5 minutes was used to melt the matrix material and to ensure a uniform temperature. After the waiting time had elapsed, a constant rate was applied and the resulting pulling force and displacement were recorded. The shearing action was stopped after around 10-mm crosshead displacement, but the force logging continued to
Figure 2: Schematic side view of the friction tester setup with a specimen for ply-ply friction (black lines). The setup was mounted in a universal testing machine. A normal force $F_n$ can be applied on the specimen in the contact area $A$ through the pressure platens (dark grey). Sliding between the plies occurs at a prescribed rate $V$, while the required pulling force $F_{\text{pull}}$ is measured.

measure the relaxation behavior and residual stress after full relaxation. The residual stress, $\tau_y$, was used to distinguish the viscoelastic matrix contribution, $\tau_{\text{mat}}$, from the total measured response,

$$\tau = \tau_y + \tau_{\text{mat}},$$

in which it is assumed that $\tau_y$ remains constant during the test, as suggested by the ply-ply friction findings of Murtagh et al. [11] on UD C/PEEK.

A range of sliding velocities (1-200 mm/min) was used with a fresh specimen for each test. Temperature was kept constant at 385 °C and 365 °C for C/PEEK and C/LM-PAEK, respectively. The normal pressure was set at 15 kPa for both materials. Each material was tested with 34 specimens.

3. Modeling

This section presents a model to predict the characteristics of the friction response. We conclude with a procedure to characterize the matrix interlayer thickness distribution in a ply-ply interface, from either micrographs or generated fiber distributions, but first further background information on wall slip is provided.
3.1. Background

Our starting point for the modeling work is the presence of a thin viscous layer of matrix material in the ply-ply interface, as frequently mentioned in various studies on inter-ply friction of TPC in melt (e.g. [4, 11, 13, 25]). A matrix interlayer was also assumed in our recent work [19], in which different hypotheses for the transient friction response, as shown in Fig. 1, were evaluated. Possible explanations for the transition from \( \tau_p \) towards the lower \( \tau_\infty \) included nonlinear viscoelasticity of the matrix material, a matrix interlayer thickness increase, or a slip relaxation effect, gradually giving rise to wall slip with time. We considered the last hypothesis as most probable, in line with earlier work of Sachs [4]. Therefore, the modeling efforts in this study focus on a wall slip mechanism.

Wall slip is generally divided into weak slip and strong slip [10, 26–31]. Weak slip relates to only small deviations of the measured shear stress from the no-slip condition due to desorption of polymer chains from the wall, or fiber in the present case, and/or disentanglement of few surface chains from the bulk ones, depending on the surface energy, above a critical shear stress \( \tau_{c,1} \).
Weak slip is said to be succeeded at higher rates by strong slip, in which adsorbed polymer chains disentangle from the bulk at a certain second critical shear stress $\tau_{c,2}$, leading to the formation of a disentangled layer near the wall [26–28]. The occurrence of strong slip is accompanied by a kink in the flow curve of shear stress versus rate, showing a shear stress plateau equal to $\tau_{c,2}$ in case of rate-controlled tests [28, 32] or a large increase in rate in case of stress or pressure-controlled experiments [26, 27, 29, 33].

Wall slip does not happen instantaneously, but evolves with time, as discussed by Hatzikiriakos and co-workers [26, 34, 35] in their research on transient wall slip effects of pure polymer melts subjected to shear flow in a sliding-plate rheometer. Delayed wall slip could occur due to a different relaxation behavior of polymer chains close to the wall with respect to the ones in the bulk [26, 29, 32, 36]. Consequently, wall slip gradually increases with the ongoing deformation, i.e. a slip relaxation effect, reducing the shear stress. Hence, the transition from no-slip towards an equilibrium slip state leads to a peak shear stress followed by a lower steady-state or long-time shear stress.

3.2. Shear flow through a ply-ply interface

A schematic illustration, representing a cross-sectional view of the ply-ply interface, is shown in the upper-left corner of Fig. 4, with fibers of two adjacent plies separated by the $x$-axis. Matrix material is assumed to be present between the fibers, resulting in a certain matrix interlayer thickness distribution, $h(x)$. Shearing in the ply-ply interface occurs when the sliding rate $V$ is applied to the top ply, as visualized in the schematic side view in the upper-right corner of Fig. 4. Hence, the matrix material will be subjected to a local shear rate $\dot{\gamma}$, depending on the applied rate and the local matrix interlayer thickness through $V/h(x)$. A 1D shear flow in the sliding direction is assumed and any interference of flow in the perpendicular or width direction is neglected. Using Equation (1), representing the matrix viscosity as function of shear rate, the local viscous shear
stress then yields:
\[ \tau_{\text{visc}}(\dot{\gamma}) = \eta(\dot{\gamma}) \dot{\gamma} \quad \text{with} \quad \dot{\gamma}(x) = \frac{V}{h(x)}. \] (4)

As the local matrix interlayer thickness changes over the width of the ply-ply interface, so will the local shear rate and thus the shear stress, as schematically illustrated in the graphs in Fig. 4. The average shear stress over the full width of the matrix interlayer, \( w \), can be calculated as:
\[ \tau_{\text{avg}} = \frac{1}{w} \int_{0}^{w} \tau_{\text{visc}}(\dot{\gamma}) \, dx, \] (5)

which we will use to predict the measured peak shear stress of the matrix contribution \( \tau_{\text{mat,p}} \).

The typical friction response shows an evolution from the peak towards the long-time shear stress, as schematically depicted in Fig. 1, which could be due to a slip relaxation effect, as mentioned earlier. Solely the effect of fully developed strong slip will be considered here, assuming that weak slip is negligible (see section 3.1). Hence, a critical shear stress for the onset of strong slip, \( \tau_{c,2} \) (following the convention in literature), is taken into account to represent the interfacial failure due to disentanglement [26–29, 33, 37]. This critical shear stress limits the local shear stresses, as visualized by the dashed grey line in the lower graph in Fig. 4. Equation (4) can be rewritten to include \( \tau_{c,2} \), resulting in a new formulation for the local viscous shear stress,
\[ \tau_{\text{visc,slip}}(\dot{\gamma}) = \min \left[ \eta(\dot{\gamma}) \dot{\gamma}, \tau_{c,2} \right]. \] (6)

The viscous term in Equation (6) will grow with rate, while \( \tau_{c,2} \) remains constant, resulting in more regions that suffer from strong slip with increasing rate. At high rates, a full-slip condition will be present, resulting in a shear stress response equal to \( \tau_{c,2} \). The value for \( \tau_{c,2} \) will be experimentally determined in this study, as it is a material parameter. Equation (6) can be combined with Equation (5) to compute the average shear stress \( \tau_{\text{avg,slip}} \) over the full width of the matrix interlayer thickness distribution with strong slip taken into account, which we will use to predict the measured long-time shear stress of the matrix contribution \( \tau_{\text{mat,\infty}} \).
Figure 4: Schematic illustration of a ply-ply interface of two adjacent plies, in which the fiber edges were searched for above and below the $x$-axis to compute the thickness of the matrix interlayer, $h(x)$, as shown in the upper graph. The top ply moves along the $z$-axis (in the fiber direction) with the applied sliding rate $V$, generating a local shear rate (visualized in the side-view in the upper-right corner) and, due to $h(x)$, a distribution of shear rates (middle graph). Consequently, when a certain viscosity is considered, a shear stress distribution appears as shown in the lower graph, in which the dashed grey line represents a critical shear stress for the onset of strong slip, $\tau_{c,2}$, limiting the local shear stresses.
3.3. Matrix interlayer thickness distribution

The matrix interlayer thickness distribution $h(x)$ forms an important input for the shear flow model described above. The local thickness is dictated by the fiber distribution near the ply-ply interface (see Fig. 4). It can be determined using image analysis by searching for the fiber edges above and below the interface. The images themselves can be obtained experimentally, using microscopy on tested specimens, or computationally by means of generated fiber distributions. We will discuss both approaches next.

3.3.1. Cross-sectional ply-ply micrographs

As a first approach, we determined the matrix interlayer thickness distribution from the cross-sectional micrographs. For this purpose, the images were first analyzed in ImageJ [38] to determine the fiber center coordinates by smoothing and evaluating the local maxima [39]. The fiber center coordinates were then loaded into Matlab, in which the relevant fiber edges in the ply-ply interface could be determined following the procedure as depicted in Fig. 4 with an assumed fiber diameter of 7 µm. Because the ply-ply interfaces between adjacent plies were hard to distinguish, we assumed that the interfaces were located at 1/3 and 2/3 of the specimen’s thickness. Note that the relevant fiber edges for the upper and lower parts of the matrix interlayer were not restricted to the baseline, as the fiber center coordinates were leading. Hence, the lower part of the matrix interlayer could be identified above the baseline when a fiber center coordinate was close to the baseline. In the rare case a fiber center coordinate coincided with the baseline, it was attributed to the upper ply.

3.3.2. Generated fiber distributions

The ply-ply interface can be mimicked using a generated fiber distribution, reducing the required experimental work and potentially enabling parameter studies on the effect of the ply-ply interface morphology on the friction response. Here, we used the algorithm as proposed by Melro et al. [40], which was kindly shared by the original authors. Random fiber distributions with a fiber diameter
of 7 µm and a fiber volume fraction of 59% were generated in a rectangle. The height of this rectangle was set equal to 10 times the fiber diameter, which was sufficient to evaluate a matrix interlayer thickness distribution at half the height or centerline. A lower rectangular height led to an unreliable result, as no fiber edges could be identified at some locations due to the fewer fibers present in the height direction, while a larger rectangular height had no significant effect on the subsequent shear-stress prediction. The width of the rectangle was limited to 50 times the fiber diameter. An increased width did not affect the subsequent shear stress prediction. Although the generated fiber distribution yields a representation of the cross-sectional core of a single UD ply, rather than a combination of adjacent UD plies, the outlined procedure can be used as well to mimic the ply-ply interface if the fiber distribution of the UD ply is sufficiently homogeneous over the full cross-sectional area, including near the ply’s surfaces.

4. Results

First, the results of the rheometry on the matrix materials will be presented, followed by our work to identify the matrix interlayer thickness distribution, $h(x)$. Thirdly, the measured ply-ply friction response of C/PEEK and C/LM-PAEK will be presented, with a particular focus on C/PEEK. This section concludes with the shear-stress predictions based on the shear flow model as described in section 3.2, using the matrix viscosities and matrix interlayer thickness distributions.

4.1. Rheometry

The viscosity curves of PEEK 150P with shear rate at different temperatures are shown in Fig. 5a. The viscosity curve at 385 °C, the same temperature as used with the friction experiments, was obtained by interpolation of the measured data and was fitted with the Cross model. The viscosity curve of LM-PAEK AE250P with shear rate at 365 °C is shown in Fig. 5b, which
was also fitted with the Cross model. Additional capillary data on LM-PAEK AE250P, interpolated between 360 and 380 °C from data kindly shared by the manufacturer Victrex [41], corresponds well with the Cross model prediction at high rates. The discrepancy at low rates probably has to do with the capillary data becoming unreliable at these rates [41]. The Cross model representations of the matrix viscosities will be used as an input for the predictive model for the peak and long-time shear-stress values.

4.2. Matrix interlayer thickness distribution

A part of a cross-sectional micrograph of a C/PEEK ply-ply specimen tested at 5 mm/min is shown in Fig. 6a. Although the cross-section consists of three UD plies, the two ply-ply interfaces are hard to distinguish as emphasized by the inset of an enlarged section of the assumed upper ply-ply interface. No clear difference was seen between micrographs of specimens tested at 5 and 25 mm/min (not shown here). A section of a micrograph of a C/LM-PAEK specimen tested at 5 mm/min is shown in Fig. 6b. In this particular section, a matrix interlayer is seen to be present between the central and upper ply as visualized by the inset at the top. However, the interface of the central ply with the bottom one is hard to identify (see lower inset), which applies to the majority of the analyzed micrographs. Although it appears that the cross-section of C/LM-PAEK has more matrix rich regions compared with C/PEEK, both materials show a fairly homogeneous fiber distribution in the ply-ply interface.

Next, we used the procedure as described in section 3.3 and schematically shown in Fig. 4 to determine the matrix interlayer thickness distribution. Fig. 7 shows a part of a C/PEEK ply-ply interface with identified fiber center locations (grey points) and the evaluated fiber edges (solid black lines) above and below the baseline (thick black line). Narrow valleys were identified by means of evaluating steep ascents and descents (¿—67.5°—). If the horizontal distance between a steep ascent and descent was smaller than the fiber diameter, the narrow valley was filled with a circular arc (dashed lines) to smooth the profile. The matrix interlayer thickness distribution was now obtained as the height
Figure 5: Viscosity curves of PEEK 150P at different temperatures (a), interpolated to obtain data at 385 °C, which was fitted with the Cross model (1) and viscosity curve of LM-PAEK AE250P (b) with Cross model fit at 365 °C together with interpolated capillary data kindly provided by Victrex [41].
4.3. Ply-ply friction tests

The marked effect of the applied rate on the ply-ply friction response of UD C/PEEK tape is shown in Fig. 8a and 8b with displacement and time, respectively. At high rates, the shear stress increased towards a maximum, then decayed followed by a steady-state response with ongoing deformation. Hence, we can distinguish a shear stress overshoot or peak, $\tau_p$, and a steady-state or long-time shear stress, $\tau_\infty$. Moreover, the relative magnitude of the peak shear stress increased with rate while the peak width reduced in time, resulting in sharper peaks as shown in Fig. 8b. The peak shear stress was not present at
Figure 7: Identified fiber center locations (grey points) from a part of a cross-sectional micrograph of a tested C/PEEK specimen. The relevant fiber edges in the ply-ply interface were evaluated (solid black lines) above and below the baseline (thick black line). Smoothening was applied (dashed lines) by means of filling narrow valleys with arcs.

low rates, as the shear stress increased monotonically towards a steady region. In that case, $\tau_p$ and $\tau_\infty$ were equal and evaluated at the first emerge of the steady region.

The subsequent relaxation behavior of C/PEEK after sliding at different rates is shown in Fig. 9. A residual stress, $\tau_y$, was consistently present after full relaxation and was evaluated after around 30 s. An average value of 1.8 kPa was found for C/PEEK, whereas the residual stress of C/LM-PAEK was a bit lower with 0.5 kPa on average. The residual stresses were used to compute the viscoelastic matrix response, $\tau_{mat}$, according to Equation (3).

The peak and long-time shear stresses of the viscoelastic matrix response, $\tau_{mat,p}$ and $\tau_{mat,\infty}$, are shown as function of the applied rate in Fig. 10 and 13 for C/PEEK and C/LM-PAEK, respectively. The flow curves show an increase of $\tau_{mat,p}$ with rate whereas $\tau_{mat,\infty}$ reaches a limit value at higher rates, ending up in a shear stress plateau with increasing rate. We will use this shear stress plateau value, 35 kPa for C/PEEK and 55 kPa for C/LM-PAEK, as the critical
Figure 8: Shear stress vs. displacement of C/PEEK with different rates (a) at a temperature of 385 °C and normal pressure of 15 kPa. A mean curve over 5 measurements conducted at 25 mm/min is shown by the grey line, where the error bars represent the sample standard deviation on the measured shear stress and (b) shear stress vs. time.
shear stress for the onset of strong slip, $\tau_{c,2}$, when we predict the flow curves in the next section.

4.4. Modeling of the peak and the long-time shear stress

The acquired matrix interlayer thickness distributions were used to model the characteristics of the transient friction response, i.e. the peak and long-time shear stress. Assuming that no slip occurs yet during the initial start-up, the local shear stress distribution for the peak values was predicted using Equation (4). Conversely, Equation (6) was used to describe the local shear stress distribution for the long-time values where wall slip is expected to occur. Then, the local shear stress distribution was averaged over the width of the matrix interlayer via Equation (5) to obtain $\tau_{\text{avg}}$ and $\tau_{\text{avg,slip}}$ for the no-slip and slip condition, respectively. Multiple matrix interlayer thickness distributions were evaluated to get a proper representation of the ply-ply interface and, therefore, the shear-stress prediction per interlayer, $\tau_{\text{avg}}$, was averaged over all predictions to obtain an average, $\bar{\tau}_{\text{avg}}$, with a sample standard deviation for the shear-stress prediction at a certain rate. The same was done for $\tau_{\text{avg,slip}}$ to yield $\bar{\tau}_{\text{avg,slip}}$. 

Figure 9: Stress relaxation curves of C/PEEK after sliding with different rates at 385 °C and 15 kPa. A residual stress, on average 1.8 kPa, remained after full relaxation.
Figure 10: Flow curve of C/PEEK measured at 385 °C and 15 kPa including predicted shear stresses using the matrix viscosity and 16 matrix interlayer thickness distributions from micrographs (solid grey line) and 16 smoothed distributions without (solid black line) and with (dashed black line) strong slip taken into account through a critical shear stress, $\tau_{c,2}$. 
The peak shear-stress prediction for C/PEEK is shown in Fig. 10 by the solid grey line for a range of sliding rates together with the measured peak shear stresses, $\tau_{\text{mat,p}}$. The prediction is based on 16 matrix interlayer thickness distributions evaluated from micrographs, with the error bars denoting the sample standard deviation. Smoothing of the matrix interlayer, by filling narrow valleys by circular arcs, results in a slightly improved prediction of the measured data at low rates as shown by the solid black line in Fig. 10. In general, a good correlation is obtained between the predicted and measured peak shear stress, $\tau_{\text{mat,p}}$, when using the matrix viscosity and matrix interlayer thickness distribution at the ply-ply interfaces.

At higher rates, the shear stress decreases after the peak to reach a steady-state region (see Fig. 8a). This decrease can also be observed in Fig. 10, which shows that the steady-state or long-time shear stress (circle symbols) deviates from the peak shear stress (triangles) at rates exceeding 0.1 mm/s. In addition, the circles in the graph show that the long-time shear stress approaches a limit value for higher rates, resulting in a shear stress plateau. Equation (6) was used to include the effect of strong slip through $\tau_{\text{c,2}}$, for which we used the measured $\tau_{\text{mat,\infty}}$-plateau value of 35 kPa for C/PEEK. The resulting flow curve prediction, again using 16 smoothed thickness distributions, is shown in Fig. 10 by the dashed black line. The matrix interlayer thickness distributions, which govern the shear stress distributions, result in a gradual transition from no-slip at low rates towards a full-slip condition at high rates. A good correlation between measurement and prediction was obtained. This good agreement can also be seen in Fig. 11, which shows the predicted versus the measured long-time shear stress at corresponding rates.

In the analysis above, the predicted peak and long-time shear stress were based on matrix interlayer thickness distributions that we derived from actual micrographs. The predicted flow curves based on matrix interlayer thickness distributions from generated fiber distributions are shown in Fig. 12, together with the experimental data on C/PEEK. The obtained predictions are comparable to the ones based on actual micrographs. The no-slip curve ($\bar{\tau}_{\text{avg}}$, solid
Figure 11: Prediction versus measurement of the long-time matrix shear stress of C/PEEK at 385 °C and 15 kPa, ranging from a nearly no-slip condition at low shear stresses towards a full-slip regime at the critical shear stress, $\tau_{c,2}$ (represented by the dashed line).

The flow curve with wall slip taken into account ($\bar{\tau}_{avg,slip}$, dashed line) also coincides well with the measured $\tau_{mat,\infty}$, again showing a smooth transition from no-slip towards a full-slip situation with increasing rate due to the distribution of thicknesses in the matrix interlayer.

We used the same procedure to predict the measured peak and long-time matrix response of C/LM-PAEK. The average shear-stress prediction based on 18 smoothed matrix interlayer thickness distributions from analyzed micrographs and the Cross model viscosity (see Fig. 5b) is shown by the solid grey line in Fig. 13, together with the experimental data. The error bars denote the sample standard deviation of the shear stress prediction at certain rates, which is larger compared with the findings on C/PEEK due to the larger variation in the matrix interlayer thickness distributions. Flow-curve predictions based on ten matrix interlayer thickness distributions from generated fiber distributions with 59% fiber volume fraction are represented by the black lines. A good shear-stress prediction is obtained for $\tau_{mat,p}$ (solid line) as well as for $\tau_{mat,\infty}$ (dashed line).
Figure 12: Predicted flow curves for C/PEEK based on the matrix viscosity and ten matrix interlayer thickness distributions from generated fiber distributions with 59% fiber volume fraction together with the measured data. Strong slip was included through a critical shear stress, $\tau_c$, which was set equal to the measured $\tau_{\text{mat, \infty}}$-plateau value of 35 kPa. If $\tau_{c,2}$ is set equal to the measured $\tau_{\text{mat, \infty}}$-plateau value of 55 kPa.

5. Discussion

The experimental and modeling results combined provide insight into the mechanisms governing the transient ply-ply friction response for UD reinforced C/PEEK and C/LM-PAEK tape. As mentioned earlier, this response consists of a static $\tau_y$ and a viscoelastic matrix $\tau_{\text{mat}}$ contribution. The latter is further characterized by a peak $\tau_{\text{mat,p}}$ and long-time shear stress $\tau_{\text{mat,\infty}}$. These three characteristics can be measured in a single experiment, as schematically illustrated in Fig. 3, and shown for C/PEEK in Fig. 8a and 9. Based on multiple measurements at different rates, we generated a flow curve of shear stress vs. rate to visualize the rate-dependency of $\tau_{\text{mat,p}}$ and $\tau_{\text{mat,\infty}}$, as shown in Fig. 10 and 13 for C/PEEK and C/LM-PAEK, respectively. The peak shear stress is governed by viscous flow of the matrix interlayer in the ply-ply interface, while...
Figure 13: C/LM-PAEK flow curve measured at 365 °C and 15 kPa including predicted shear stresses using the matrix viscosity and 18 smoothed matrix interlayer thickness distributions from micrographs (solid grey line). The prediction based on ten matrix interlayer thickness distributions from generated fiber distributions with 59% fiber volume fraction is shown with no-slip condition ($\bar{\tau}_{\text{avg}}$, solid black line) and with strong slip included ($\bar{\tau}_{\text{avg,slip}}$, dashed black line).
the long-time shear stress exhibits wall slip, as shown by the successful application of a microscopic shear flow model. In this section, we will discuss the obtained results, as well as the underlying mechanisms, by addressing the characteristic shear stress values that define the friction response. Where possible, we will relate the current findings to earlier observations found in literature.

5.1. Static contribution \( \tau_y \)

The relaxation curves of the measured friction response showed that a static contribution \( \tau_y \) of approximately 1.8 kPa and 0.5 kPa was present after full relaxation for C/PEEK and C/LM-PAEK, respectively. These values correlate well with data reported in literature. Murtagh et al. [11], for example, performed force-controlled friction experiments on UD C/PEEK and found that a minimal shear stress, which they denoted as a yield stress, of approximately 2 kPa was required to induce irreversible ply-ply slippage. Moreover, ply slippage ceased when the shear stress dropped below this threshold value, which substantiates our assumption of a static contribution. A comparable small yield stress was also found for other material systems, such as for UD C/PP [42], and with other experimental approaches, such as oscillating rheometry on UD C/PEEK leading to a yield stress of approximately 1 kPa [43, 44]. The data of Deignan et al. [45] on intra-ply shear of C/PEEK also suggest a rate-independent contribution to the shear stress, which was labeled as Coulomb friction in a later study [46]. A Coulomb friction or Bingham plastic model is frequently used in combination with a power law to fit the experimental friction data [7, 17, 25, 47], implicitly assuming a static shear stress contribution throughout the measurement, inline with this study. As a possible explanation, several authors suggested that the yield stress could originate from fiber-fiber friction [11, 25, 48].

5.2. Peak shear stress \( \tau_{\text{mat.p}} \)

The model that we proposed accurately predicts the peak shear stress for C/PEEK using the matrix viscosity and the matrix interlayer thickness distribution (see Fig. 10), as obtained from cross-sectional micrographs of tested
specimens. This result shows that the peak shear stress is the product of the 1D shear deformation of a purely viscous polymer with a nonuniform thickness distribution and that 2D and viscoelastic effects can be neglected. The prediction for C/LM-PAEK, again using micrographs to obtain the matrix interlayer thickness distribution, slightly exceeds the measured data, as is shown in Fig. 13. In addition, the error bars indicate a large variation in the predicted shear stress. A possible explanation can be found in the cross-sectional micrographs of the C/LM-PAEK plies, as provided in Fig. 6, which show a slightly less homogeneous fiber distribution with more matrix-rich regions compared to the C/PEEK plies. Hence, a variation in the acquired matrix interlayer thickness distribution is to be expected, leading to a larger variation in the shear stress prediction. The slightly higher local matrix content in the interface may also explain the lower yield stress for C/LM-PAEK relaxation compared with C/PEEK.

Generated fiber distributions were used to circumvent the issue of identifying the ply-ply interface from the micrographs. Such an approach is warranted in the case of a (nearly) homogeneous fiber distribution in the ply-ply interface, which seems applicable for the current research considering the micrographs in Fig. 6 and the good correspondence of the resulting shear stress predictions with the measured peak shear stress for both C/PEEK and C/LM-PAEK, as shown in Fig. 12 and 13, respectively.

5.3. Long-time shear stress $\tau_{\text{mat,}\infty}$

We considered that the decrease in shear stress after $\tau_{\text{mat,p}}$ is caused by a gradual increase of wall slip. As mentioned earlier, two modes for wall slip are generally distinguished, namely weak slip and strong slip [10, 26–31]. As the former only results in small deviations compared to the no-slip condition, we only considered the effect of strong slip. Here, a disentangled layer is formed near the wall at a critical shear stress $\tau_{c,2}$, which we used to limit the local viscous shear stresses in the interlayer. The thickness distribution in the interlayer then determines the rate-dependency of the long-time shear stress $\tau_{\text{mat,}\infty}$. This
dependency can be explained by considering the shear stress distribution $\tau(x)$, which is directly influenced by the thickness distribution $h(x)$ as illustrated in Fig. 4. At a certain rate, the local viscous shear stress may exceed the critical shear stress $\tau_{c,2}$ resulting in local slip. As the rate increases, the viscous shear stresses increase, causing a larger fraction of the interface to experience slip. Hence, a gradual transition from no-slip to full slip with increasing rate is observed. This transition, and thus also the shape of the predicted flow curve, is determined by the matrix interlayer thickness distribution. The application of a constant critical shear stress, independent of the local matrix interlayer thickness, is warranted based on the findings of Wang and co-workers [27, 33], who showed that $\tau_{c,2}$ remains constant irrespective of the gap height used in their sliding plate experiments on an entangled polymer melt.

The effect of strong slip, together with the matrix viscosity and the matrix interlayer thickness distribution, can be used to accurately predict the measured long-time shear stress values $\tau_{\text{mat,}\infty}$ for both C/PEEK and C/LM-PAEK. However, in practice, strong slip will gradually develop during the friction measurement, as disentangling cannot happen instantaneously. As such, a slip relaxation or delayed slip effect occurs [26, 34, 35]. The good agreement between the measured peak shear stress $\tau_{\text{mat,p}}$ and the predicted shear stress using a no-slip condition, as shown in the flow curves in Fig. 10 and 13, suggest that wall slip is negligible during start-up. The slight overestimation of the measured peak shear stress at the highest rates, though, could suggest that partial slip takes place before the peak shear stress is reached for these rates. Additional experiments at higher rates, possibly in combination with a model that describes the evolution of slip with time, are required to confirm this.

5.4. Critical shear stress $\tau_{c,2}$

Strong slip occurs when a critical shear stress $\tau_{c,2}$ is achieved. As a first step, we have set the critical shear stress equal to the measured $\tau_{\text{mat,}\infty}$-plateau values in the flow curves. The rate-independence of the long-time shear stress at these high rates suggests that the deformation is fully governed by slip rather
than viscous flow. In addition, almost all of the simulated local shear stresses in the matrix interlayer exceeded the measured macroscopic shear stress at these high rates.

We found a critical shear stress of 35 kPa and 55 kPa for C/PEEK and C/LM-PAEK, respectively. Several studies discussed wall slip for a range of different polymers [26, 27, 32, 34, 35, 49], in particular related to extrusion problems [28, 30, 31, 37, 50], and mention critical shear stresses in the range of 100 to 400 kPa. The main difference with our work is the interface between polymer chains and carbon fibers, while the cited research mostly considered the more common metal-polymer interfaces. The high surface energy of metals [27, 28] compared with carbon and the different polymers considered might explain why the values in literature are a bit higher. Moreover, pressure-driven devices, such as a capillary rheometer, result in higher slip stresses than those obtained with rate-controlled equipment, such as a sliding-plate rheometer. Boukany et al. [33] for example reported an increase of the critical shear stress for polybutadiene melt from 200 kPa, based on sliding-plate rheometry, to 300 kPa when using a pressure-driven setup. Finally, Adewale and Leonov [37] mentioned that the critical stress “as observed in many experiments ranges” from 10 to 1000 kPa. Although the critical stress values obtained in this research correspond with this broad range, future work is to further quantify the magnitude of $\tau_c$, for ply-ply friction under different conditions by means of flow curves at different temperatures and normal pressures.

The application of a critical shear stress, thus taking strong slip into account, leads to a better understanding of ply-ply friction, as the results substantiate the concept of wall slip as the underlying mechanism responsible for the observed transient response. Although the current research addressed only 0°-0° interfaces, the principles seem applicable to other ply orientations as long as the ply-ply interface can be determined accurately. Future work is to measure and model this orientation effect. Further, the proposed predictive model for the peak shear stresses, based on the matrix interlayer thickness distribution and the matrix viscosity, combined with a critical shear stress for the long-time shear...
stress response, sets the basis for future constitutive modeling. A transient description, relating the peak and long-time shear stress, is needed to improve the predictive capabilities of forming simulation software.

6. Conclusion

We investigated the transient ply-ply friction response of UD C/PEEK and C/LM-PAEK through a combination of experimental and modeling work. The measured shear stress was characterized by a static and a viscoelastic matrix contribution, exhibiting a peak followed by a steady-state or long-time shear stress. The peak shear stress was successfully modeled based on the matrix viscosity and the matrix interlayer thickness distribution in the ply-ply interface, which was analyzed from actual cross-sectional micrographs of tested specimens as well as from generated fiber distributions.

A slip relaxation effect, gradually giving rise to wall slip, explains the observed transition from the peak towards the long-time shear stress. We modeled the effect of wall slip on a microscopic level through a critical shear stress for the onset of strong slip, which resulted in an accurate prediction of the long-time values. A gradual transition of the macroscopic shear stress was obtained from no-slip towards full-slip with increasing rate, a phenomenon caused by the inhomogeneous matrix interlayer thickness in the ply-ply interface. Thus, the measured characteristics of the transient friction response can be predicted accurately solely based on the matrix viscosity and the matrix interlayer thickness distribution in the ply-ply interface when accounted for strong slip by means of a critical shear stress. This result reduces the number of tests required to characterize a material. Moreover, this finding substantiates the concept of wall slip as a key underlying mechanism for the transient ply-ply friction response, which will form the basis for future constitutive modeling of ply-ply friction to improve the predictive capabilities of process simulation software on defect generation in hot press forming.
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References


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Highlights of ‘Prediction of the peak and steady-state ply-ply friction response for UD C/PAEK tapes’:

- Ply-ply friction tests were performed on C/PAEK UD tapes for a wide range of rates
- The peak friction increases with rate, while the steady-state reaches a plateau
- The peak can be predicted from the interface thickness distribution and viscosity
- The steady-state friction can be described using the concept of wall slip
- The gradual increase of wall slip explains the observed start-up friction response
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Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: