

# On the Effect of Release Agent and Heating Time on Tool-Ply Friction of Thermoplastic Composite in Melt

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**Abstract.** Process simulation software for hot press forming requires accurate material characterization. One of these characterization experiments concerns tool-ply friction, for which the methodology is well established. However, the experimental conditions are often not representative for the actual forming process. This research focuses on the effect of release agent and heating time on the tool-ply friction response. UD carbon fiber-reinforced PEKK was forced to slide against metal foils in a benchmarked friction tester at different rates, normal pressures and temperatures. The typical friction response, exhibiting a shear stress overshoot followed by a steady-state region, did not qualitatively change when applying a Marbocote 227CEE release agent on the metal foils. However, the overshoot reduced and, in case of a high normal pressure of 45 kPa, the steady-state response lowered as well. Thus, release agent should be included for a more accurate characterization of tool-ply friction. A longer heating time resulted in a large increase of the overshoot, whereas the steady-state response was nearly unaffected. The same observation was made when testing at a higher temperature, which may suggest that the increase in overshoot is due to increased adhesive bonding. Moreover, a change in adhesive bonding could also explain the lower overshoot observed when a release agent was applied, indicating adhesion as a key mechanism for tool-ply friction.

## Introduction

Hot press forming of continuous fiber-reinforced thermoplastics is an attractive technology to manufacture lightweight structural parts in high volumes. However, the use of hot press forming can be challenging, especially, for complex parts with double curvature, which may suffer from forming induced defects. The presence of these defects, like wrinkles, results in weak spots and, therefore, part rejection [1]. Especially uni-directional (UD) reinforcements are more susceptible to forming induced defects compared with fabric reinforcements [2, 3].

The time and costs involved to solve forming problems can be minimized by using process simulation software beforehand, facilitating the design stage. Obviously, the predictions of these process simulation tools then need to be accurate, especially on defect generation, in order to truly enable first-time-right defect-free manufacturing. In turn, accurate predictions require proper material characterization. Hence, it is of utmost importance to accurately characterize the formability of a material to obtain appropriate constitutive relations for the use in simulations.

One of the properties to be characterized is the resistance against movement between tooling and surface plies or tool-ply friction [1]. Evidently, the experimental conditions when characterizing tool-ply friction should be representative for the actual press forming process. Hence, release agents that are used to help part removal after forming should be accounted for [4], which is often not the case. Murtagh et al. [5] did include release agents in their study on tool-ply friction of UD C/PEEK, but only

reported on the steady-state response. To the best of our knowledge, the influence of release agent on the full transient tool-ply friction response has not been considered in open literature yet.

The contact or heating time prior to sliding is another parameter that is typically different in testing compared to practice. In the experimental characterization, ply and tooling surface are often already in contact prior to sliding to facilitate heating, whereas tool-ply sliding occurs often immediately when contact has been formed in the actual process [6]. Although an experimental set-up featuring a separate specimen pre-heating stage was proposed to circumvent the issue of contact heating [7], most characterizations are performed on set-ups without this feature to guarantee an accurate and homogeneous temperature distribution. Hence, most researchers used a constant heating time, without reflecting on the effect of this choice [1, 8], or did not explicitly report on the value used [5, 9, 10].

The effect of release agent and heating time on the transient (start-up) tool-ply friction response of UD fiber-reinforced PEKK in melt was investigated in this work. These findings will help to improve characterizations on tool-ply friction for the purpose of simulation software. In the end, improved predictions on defect generation will in particular help industry to produce first-time-right defect-free TPC parts by means of hot press forming and as such be instrumental to further increase the use of lightweight TPC parts.

## Materials and Methods

**Materials.** The pre-impregnated composite material investigated in this work is known as APC (PEKK-FC), consisting of a thermoplastic matrix of PEKK reinforced with aligned continuous carbon fibers. The tape has a fiber volume fraction of 59% and a thickness of 145  $\mu\text{m}$ . The glass transition and melting temperature are 159  $^{\circ}\text{C}$  and 337  $^{\circ}\text{C}$ , respectively, according to the manufacturer Solvay. Metal foil, Martin M-Tech mild steel DC01 50  $\mu\text{m}$ , was used to represent the tooling surface, which was cleaned with isopropanol. Details on the topology of the metal surface can be found in Sachs [1]. Marbocote 227CEE was chosen as a suitable release agent, which was applied according to the manufacturer's recommendation and left to cure for 24 hours at room temperature [4].

**Experimental set-up.** The friction between the APC and the metal foil was measured by means of a benchmarked friction tester [11], schematically shown in Fig. 1. The friction tester was placed in a universal testing machine. A composite tape was mounted in the upper clamp whereas two metal foils at each side of the tape were clamped at the bottom, resulting in two tool-ply interfaces. Alignment was checked using a laser leveler. A constant rate was applied to the upper crosshead and the required pulling force,  $F_{\text{pull}}$ , was recorded using a 1 kN loadcell. The pulling force was used to compute the shear stress per slip interface,

$$\tau = \frac{F_{\text{pull}}}{2A}, \quad (1)$$

with  $A$  the contact area of 50x50  $\text{mm}^2$ , in which the temperature and pressure were controlled through heated pressure plates. An additional overlap between the APC tape and metal foils of 15 mm was used to provide a constant pressurized area during the test. The reader is kindly referred to Sachs [1] and Ten Thije et. al. [12] for more detail on the friction tester.

**Experimental conditions.** Friction tests with and without Marbocote 227CEE were performed to investigate the effect of release agent on the tool-ply friction response of APC. The response for a rate of 25 mm/min, a pressure of 15 kPa and a temperature of 385  $^{\circ}\text{C}$  was used as a reference. From the reference point, variations in the rate (5 and 125 mm/min), pressure (5 and 45 kPa) and temperature (370 and 400  $^{\circ}\text{C}$ ) were separately tested in triplicate.

A heating time of 3 minutes was used to melt the matrix material before starting the experiments. In a second series of experiments, this heating time was varied to investigate the effect on the subsequent friction response at the reference conditions (25 mm/min, 15 kPa and 385  $^{\circ}\text{C}$ ). To broaden the scope

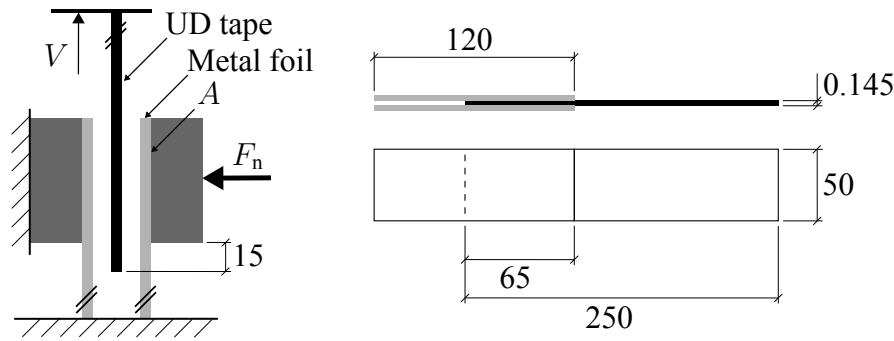


Fig. 1: Schematic overview of the friction tester, placed within a universal testing machine, with a schematic tool-ply specimen used to measure the friction response. Dimensions in mm.

of this research, ply-ply specimens, with plies surrounding the central ply instead of metal foils, were also subjected to heating times of 5 and 10 minutes at the reference conditions.

## Results

The experimentally obtained tool-ply friction response of APC will be presented in this section. The effect of a Marbocote 227CEE coating on the metal foils will be shown for different rates and pressures. Further, the effect of temperature will be presented and this section will conclude with the friction response for various heating times for both tool-ply and ply-ply friction.

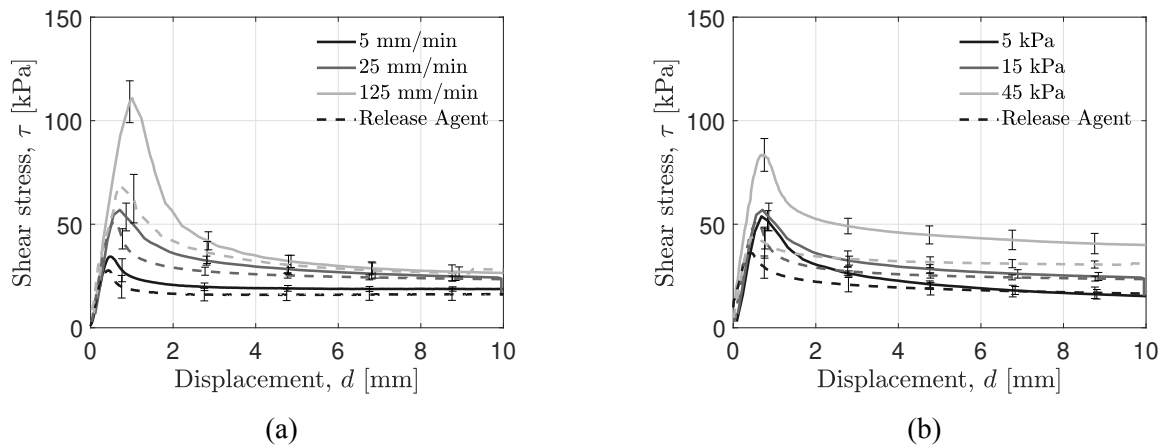


Fig. 2: Tool-ply friction response of APC with displacement for different rates (a) and normal pressures (b). Solid lines represent the response for bare metal foils and the dashed lines show the effect of coating the metal foils with Marbocote 227CEE.

**Effect of release agent.** The tool-ply friction response of APC with displacement for different rates and pressures is shown in Fig. 2a and 2b, respectively. While varying one of the variables, the others were kept constant at the reference conditions (i.e. a rate of 25 mm/min, a pressure of 15 kPa and a temperature of 385 °C). The solid lines represent the friction response for bare metal foils, whereas the dashed lines correspond to Marbocote 227CEE coated metal foils. Measurements were performed in triplicate and the average response is shown with the error bars denoting the sample standard deviation.

A start-up response can be observed, in particular when the rate was varied as shown in Fig. 2a. The shear stress increases towards a maximum or overshoot, then starts to decrease, followed by a steady-state region. This typical behavior can be observed for samples with and without release agent, although the magnitude of the overshoot is reduced when coated metal foils were tested. In contrast to the overshoot, the steady-state response seems unaffected.

An interesting observation can be made when inspecting the response for different pressures as shown in Fig. 2b. The typical friction behavior can be recognized, as well as the effect of release

agent resulting in a lower shear stress overshoot with a seemingly unaffected steady-state response. However, the steady-state response at the highest pressure measured, 45 kPa, is affected by the release agent coating, as the shear stress is lower compared to the one measured with bare metal foils.

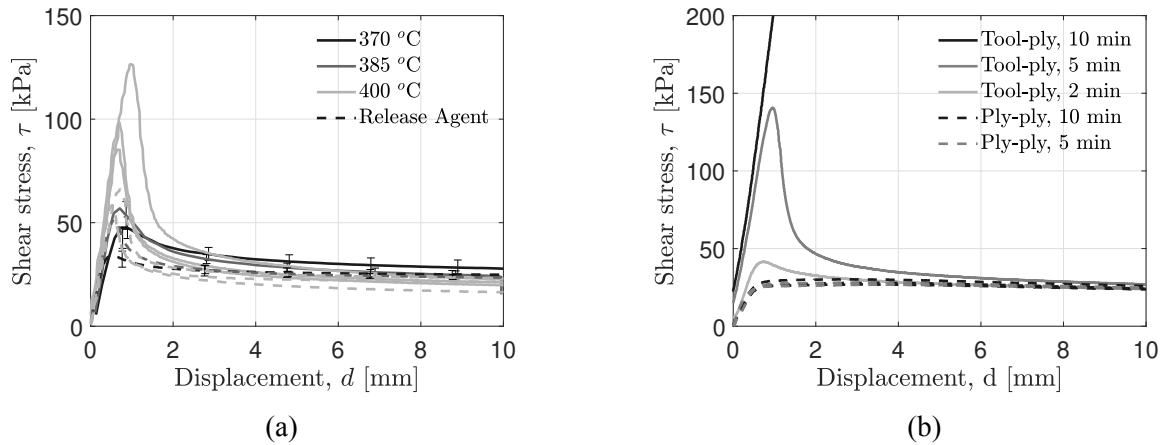


Fig. 3: Tool-ply friction response of APC at 370 and 385 °C (average responses with the error bars denoting the sample standard deviation) and 400 °C (individual tests) with and without release agent applied (dashed and solid lines, respectively) (a) and the effect of heating time on the tool-ply (solid lines) and ply-ply (dashed lines) friction response at reference conditions (25 mm/min, 15 kPa and 385 °C) (b).

The effect of temperature on the friction response for both bare and coated metal foils is shown in Fig. 3a. Average friction responses were computed at 370 and 385 °C with the error bars denoting the sample standard deviation. The individual curves are shown for the measurements at 400 °C instead of an average curve, as a relative large variation in the friction response was obtained. The typical transient friction response as previously described can also be recognized in Fig. 3a. Interestingly, the overshoots at an elevated temperature of 400 °C are considerably larger than the ones at a lower temperature. The effect of a release agent coating on the metal foils is shown by the dashed lines, whereas the tests with bare metal foils are shown by the solid lines. The overshoot is again lower if release agents are applied to the metal foils, whereas the steady-state response seems barely affected.

**Effect of heating time.** The first series of experiments used a constant heating time of 3 minutes in order to melt the matrix material. As the heating time is actually an artifact from the experimental method, the effect of this heating time on the tool-ply friction response was further explored. The friction response for a heating time of 2, 5 and 10 minutes is shown in Fig. 3b. The solid lines represent the tests with a tool-ply configuration, in which the experiment with a heating time of 10 minutes needed to be aborted as the limit of the loadcell was almost reached. A shorter heating time resulted in a lower overshoot, while the steady-state response was unaffected.

Experiments with ply-ply specimens, in which the slip interfaces are formed with adjacent plies instead of a ply-metal contact, are shown by the dashed lines in Fig. 3b. Contrary to tool-ply friction, the heating time did not significantly change the shear stress overshoot nor the steady-state response. Interestingly, the steady-state shear stress of tool-ply and ply-ply configurations was approximately equal.

## Discussion

The experimental results as presented above will be discussed in this section. The findings will be related to earlier literature where applicable and some concluding remarks will be made regarding future work.

**Effect of rate, pressure and temperature.** The tool-ply friction results of molten APC in contact with bare and Marbocote 227CEE coated metal foils, as shown in Fig. 2 and 3a, exhibit a typical response in which the shear stress increases towards a maximum, starts to decay and gradually reaches a steady-state value. Hence, an overshoot and steady-state shear stress can be distinguished. A similar response was frequently observed in earlier studies on friction of composites in the molten state (e.g. [1, 5, 8, 9, 10, 13, 14]). Apart from the qualitative similarity of the friction response with literature, the magnitude of the friction values measured in this research are comparable to the findings of Sachs [1] on tool-ply friction of UD C/PEEK.

Fig. 2 shows an increase of the overall friction response with rate and normal pressure, as expected from earlier findings [1]. Especially the overshoot increases with rate, denoting a stronger transient response, which was also observed in other studies [9, 10]. The subsequent steady-state response also increases with rate, in agreement with the characteristics of hydrodynamic lubrication of a thin matrix-rich interlayer, as often used to describe tool-ply friction [1, 9, 15, 16]. However, the marginal increase between the 25 and 125 mm/min indicates that the steady-state values tend to level off. Sachs [1] found indeed that the steady-state response reaches a limit value at higher rates for tool-ply friction of UD C/PEEK, which was later substantiated by ply-ply friction results in earlier work of the current authors [17]. Thus, the steady-state values increase with rate up to a certain critical shear stress, at which the steady-state response becomes independent of rate. Hence, the steady-state response cannot fully be described with the concept of hydrodynamic lubrication [1].

A higher temperature resulted in an increase of the shear stress overshoot as shown in Fig. 3a, which deviates from the general consensus [13]. One would expect a lower resistance at higher temperatures due to a reduction in viscosity, though the opposite could occur as a result of polymer degradation, leading to a viscosity increase. Polymer degradation, however, would probably affect the full friction response rather than solely the overshoot. A few studies also found an increase in friction with temperature. Lebrun et al. [18] measured higher tool-ply friction with temperature for woven G/PP and suggested that a reduction of the matrix-rich layer could be the cause. Murtagh et al. [5, 6] found a tool-ply friction increase with temperature for UD C/PEEK material and suggested this response to be due to failure of adhesive bonds between the matrix and tooling surface. The degree of adhesive bonding might increase due to the lower viscosity at higher temperatures, resulting in an increase of the shear resistance. The results in Fig. 3a seem to agree with the suggestion of Murtagh et al. [5], as the higher temperature particularly affects the overshoot rather than the subsequent steady-state response.

**Effect of release agent.** The effect of adding Marbocote 227CEE release agent on the metal foil's surface was most pronounced on the overshoot as shown in Fig. 2 and 3a. The typical friction response remains present when using coated metal foils. However, the overshoots are reduced, indicating that tool-ply slip is promoted in the actual process under influence of a release agent. Hence, release agents are important to consider for an accurate description of tool-ply friction in simulation software. Contrary to the overshoot, the steady-state response remains unchanged. Except for the response at a high normal pressure of 45 kPa (see Fig. 2b), as the steady-state response of bare metal foils with APC was slightly higher than the one for coated metal foils at this normal pressure. A possible explanation for the affected steady-state response at higher normal pressure can be derived from earlier findings of Sachs [1] and the current authors [17]. The experimental work of Sachs showed that an increase of normal pressure leads to an increase of the critical shear stress at which the steady-state response becomes independent of the applied rate. It was suggested that the presence of this critical point could be due to wall slip, which could arise as a result of either adhesive failure, in line with the suggestion of Murtagh et al. [5] on the overshoot, or disentangling of polymer chains near the fibers or metal foils [19]. Higher pressures are thought to postpone the occurrence of full wall slip to higher shear stresses, and thus higher rates. At rates below the critical point partial wall slip may occur, whereas a full slip condition might be present at rates above the critical point [17]. The partial slip region could be susceptible to the surface condition through the degree of adhesive failure, resulting in more or less

slip depending on the treatment. The surface conditions in the full slip regime are of less importance as full interfacial failure is expected to occur through desorption or disentangling [20, 21]. Hence, at higher normal pressures, the increased critical shear stress results in a broader partial slip region, which might explain the affected steady-state response by the surface conditions. More details can be found in Pierik et al. [17]. To further explore the probability of this slip mechanism, full flow curves of shear stress versus rate need to be measured at higher normal pressures to check under which conditions a full slip regime is present.

The influence of release agents on the tool-ply friction response of UD C/PEEK was studied by Murtagh et al. [5], who reported on the steady-state response and found a decrease in shear stress when applying release agents. Such an observation cannot be made in the current research, except for the measurements conducted at the highest normal pressure of 45 kPa. However, the measurements of Murtagh et al. [5] were performed at an even higher normal pressure of 100 kPa, which might explain why they found a stronger effect of release agents on the steady-state response compared with the current study. Firstly, a higher normal pressure results in an increased shear stress and, consequently, leads to larger shear stress differences between the conditions. Secondly, as described above, the steady-state response might be in a full slip region at low normal pressures resulting in a steady-state shear stress equal to the critical shear stress, whereas at higher normal pressures the critical shear stress increases leading to an expanded partial slip region towards higher rates. Since the partial slip region is expected to be more susceptible to the surface conditions, a larger difference in the steady-state response could be observed in the work of Murtagh et al. [5] compared with the current study.

**Effect of heating time.** The second series of test concerned the effect of heating time. A certain heating time is required to melt the matrix material before the friction test can be executed, during which contact is established between the APC ply and the metal foils. The effect of this artificial heating time on the subsequent tool-ply friction response was reported in Fig. 3b. A large increase in overshoot with heating time can be observed, whereas the steady-state responses tend to coincide. Experiments with a ply-ply configuration, in which the slip interface is formed by adjacent plies, resulted in a monotonic increase of the shear stress towards the steady-state response. The strong effect of the heating time on the overshoot was not observed, as shown in Fig. 3b. Hence, degradation of the polymer material or squeeze flow in the slip interface during the heating time seems improbable, as it is to be expected that these effects would also influence the ply-ply friction response.

One might speculate that the longer heating time resulted in an increased adhesive bonding between the APC ply and the metal foils, perhaps due to adsorption of the polymer chains to the metal surface, in agreement with the suggestion of adhesive bond creation during heat up mentioned by Murtagh et al. [5] and the observed increase in friction at higher temperatures (see Fig. 3a). Hence, there could be a sign of a time-temperature relation for the increase in adhesion. Moreover, the overshoot decrease when using a release agent might also be attributed to a change in adhesion through less strong or fewer bonds. The presented results suggest tool-ply adhesion as a key mechanism to explain the transient tool-ply friction response. In addition, the applied heat could also affect the metal foils' surface itself through oxidization with time, which might promote mechanical interlocking or improve the bond between adhered polymer chains near the wall. Future work is to measure the effect of the heating time on the friction response when using heat treated and release agent coated metal foils. Moreover, the response for other TPC tapes should be measured as well.

## Conclusions

Tool-ply friction tests, in which a molten APC tape was forced to slide against metal foils, showed a typical friction response as commonly observed in earlier studies: shear stress growth, followed by a maximum or overshoot leading towards a steady-state region. The ratio between overshoot and steady-state shear stress increased with sliding rate, whereas a relatively strong increase of the steady-state response was observed with increasing normal pressure. The presence of a release agent on the metal foils' surface did not qualitatively change the response. However, the overshoots were reduced

in general, as well as the steady-state response in case of a high normal pressure of 45 kPa in particular. Hence, release agent should be taken into account to increase the accuracy of the description of tool-ply friction in simulation software.

The characterization methodology for tool-ply friction requires a certain heating time, during which contact forms between the ply and the metal foils, whereas tool-ply friction often occurs instantaneously in the actual forming process. An increase of this artificial heating time resulted in a higher overshoot. It was argued that polymer degradation and squeeze flow are less probable reasons for the increased overshoot as ply-ply friction results were not affected. However, the higher friction observed could be due to an increased adhesion between the polymer melt and the metal foils. In the same way, an increase of the adhesive bonding might explain the larger overshoot observed at a high temperature. Further, the presence of a release agent could reduce the adhesion between the polymer melt and the coated metal foils, explaining the lower overshoot observed. Hence, adhesion seems to play a key role in the characterization of tool-ply friction.

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