Tool-ply interaction in the formation of waviness during C/PEEK consolidation

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A B S T R A C T

It is well established that fiber waviness in continuous fiber-reinforced polymer composites is a defect rather than an aesthetic flaw, as it potentially leads to a severe knockdown of the mechanical properties. Consequently, it is desirable to minimize or prevent fiber waviness during processing. This research explored experimentally the influence of tool material, release media and laminate size on the formation of in-plane fiber waviness during C/PEEK consolidation. The formed waviness was quantified and correlated to the measured tool-ply friction coefficients and thermal coefficients of expansion of the tool plates. This research provides additional evidence that tool shrinkage is the driving force for waviness formation in C/PEEK laminate consolidation. Furthermore, the formation of waviness depends on the balance between the coefficient of thermal expansion of the tool, the tool-ply coefficient of friction and the laminate size.

1. Introduction

The use of thermoplastic composites (TPC) is steadily increasing in the aerospace industry because of their potential for cost-effective, automated and rapid manufacturing. These materials entail a high specific stiffness and strength, which are natural properties of fiber-reinforced composites in general. Additionally, an increased toughness and indefinite shelf-life are inherent to the thermoplastic matrix. However, these benefits may be impaired by processing defects such as fiber waviness, which are occasionally found in parts in the industry. It is well established that fiber waviness in continuous fiber-reinforced polymer composites is a defect rather than an aesthetic flaw, as it potentially leads to a severe knockdown of the mechanical properties. Consequently, it is desirable to minimize or prevent fiber waviness during processing. This research explored experimentally the influence of tool material, release media and laminate size on the formation of in-plane fiber waviness during C/PEEK consolidation. The formed waviness was quantified and correlated to the measured tool-ply friction coefficients and thermal coefficients of expansion of the tool plates. This research provides additional evidence that tool shrinkage is the driving force for waviness formation in C/PEEK laminate consolidation. Furthermore, the formation of waviness depends on the balance between the coefficient of thermal expansion of the tool, the tool-ply coefficient of friction and the laminate size.

The most detailed research regarding the formation of waviness in TPC laminate consolidation considered the amorphous carbon fiber/polyetherketon (C/PSU) material system. It was shown that the use of graphite and Invar tool plates yielded no waviness, whereas steel plates produced modest waviness and aluminum and brass plates produced high levels of waviness [6]. In related research, the role of eight processing parameters in the consolidation of C/PSU laminates was investigated, of which only three affected the development of fiber waviness: laminate size, cooling rate, and tool plate material [7]. The two cited investigations suggest that the processing temperature is not an important parameter. However, previous research by the authors of this paper indicated that processing temperature is in fact a key parameter in the formation of waviness during consolidation of semi-crystalline C/PEEK laminates [8]. The amount of tool shrinkage during cooling, which was governed by the processing temperature, dictated the misalignment angle of the wavy fibers. The differences in relevant processing parameters between these studies were attributed to the different solidification mechanisms between the amorphous C/PSU and semi-crystalline C/PEEK material systems [8]. As a common denominator, all investigations concluded that a mismatch in shrinkage between the tool and the ply is the governing mechanism for waviness formation during laminate consolidation.

The mismatch in the coefficient of thermal expansion (CTE) between
the tool and the laminate is a well-known mechanism responsible for shape distortions in thermoset composite processing, most notably mold stretching [9]. The laminate is stretched by the expanding mold during heating, followed by the isothermal curing process where the generated stresses are frozen in. These frozen-in stresses may cause shape distortions after cooling to room temperature. Conversely, a tool-ply CTE mismatch in TPC processing has very different implications, most notably the formation of fiber waviness. For C/PEEK consolidation it was demonstrated that tool shrinkage induced fiber waviness is only formed in a specific temperature interval, designated the fiber waviness formation temperature interval \( \Delta T_w \) and defined as the difference between processing temperature \( T_{\text{max}} \) and the crystallization temperature \( T_{\text{cry}} \) [8]. The matrix has not yet solidified in this interval and cannot fully support the fibers in the transverse direction, effectively lowering the buckling resistance of the fibers, thus facilitating the formation of waviness. Frictional forces between the tool and the ply are responsible for the transfer of tool displacements to the fibers [6].

The available literature regarding the tool-ply interaction during TPC laminate consolidation is scarce. Research that did focus on friction during TPC processing mainly concerned the forming process, and focussed on ply-ply friction [10,11]. However, a few investigations do provide an insight into tool-ply friction during laminate consolidation. High coefficients of friction (CoF) were found between a steel tool and a polyimide film at processing conditions for the double diaphragm forming process [12]. A high CoF at processing conditions was also found between a unidirectional C/PEEK ply and a steel tool treated with release agents [13], along with dependencies on shear rate, temperature, normal force and fiber orientation. These dependencies were also found for woven TPOs [14,15]. However, the aforementioned investigations in this paragraph focussed on conditions during the forming process, which is a very dynamic process involving higher shear rates and a less homogeneous temperature and normal force distribution, compared with laminate consolidation. Some of those experiments did consider a low shear rate or pull-out velocity and may still provide an insight into the effective tool-ply friction during laminate consolidation. Tool-ply friction in the laminate consolidation process has only been studied qualitatively and considered the C/PSU material system: pull-out of a ply between tool plates was more difficult at processing temperatures than at room temperature [6]. Quantitative measurements were not performed in the aforementioned study. Hence, those authors recommended studying the tool-ply interaction, and its relation to waviness formation during consolidation, in greater detail. The influence of tool material and release media on the formation of waviness during consolidation of C/PEEK flat laminates are further investigated in this paper.

The occurrence of waviness in TPC parts can be reduced with a better understanding of the manufacturing process. Clear processing guidelines can be determined once the relevant aspects of the process are fully analyzed. The mechanisms governing waviness formation were already identified in previous research along with the waviness formation temperature interval [8]. However, the effect of tool material and tool-ply friction needs to be quantified for full control of waviness formation during the manufacturing process.

The main objective for this research effort is to systematically quantify the influence of processing variables on the formation of fiber waviness during consolidation of C/PEEK composite laminates. This is done by firstly determining the degree of waviness for various tool materials and release media using an experimental consolidation setup. The following variables are investigated here:

- Coefficient of thermal expansion of the tool material
- Friction between the tool and the composite ply, using various release media
- Size of the laminate in the fiber direction

Secondly, representative values for the tool-ply friction at processing conditions are determined experimentally using a friction testing setup. The findings will be used to provide processing guidelines for the production of waviness-free TPC laminates.

2. Experimental work

Single plies were consolidated to investigate the effect of the aforementioned variables on the formation of waviness during the consolidation process. Four different tool materials were used to study the effect of tool-ply CTE mismatch and four different laminate sizes were considered to study their effect on waviness formation. The tool-ply CoFs were varied using two types of release media. Release agents are typically designed for a low tool-ply friction [16]. However, relevant tool-ply friction data for the considered material system and release media were not available in the literature. Therefore, the CoFs were determined using a friction testing setup. The degree of waviness in the laminates was determined with optical surface microscopy and a fiber tracing algorithm.

2.1. Single-ply consolidation experiments

2.1.1. Vacuum-assisted oven consolidation setup

The setup shown in Fig. 1 was used to consolidate single C/PEEK composite plies. A 300 mm × 300 mm ply was stacked between release media and tool plates, while pressure and heat were applied to the stack by a vacuum bag inside a convection oven. Previous work indicated that this type of setup is suitable for studying the formation of waviness [8]. The temperature was measured by two thermocouples, placed not between, but right next to the tool plate, to avoid any disturbance in the ply thickness. The graphite sheet provides a slip plane between the base plate and the tool plate. This slip plane ensures that the thermal expansion of the tool plate is not constrained by the base plate. Details about the parts in this setup are shown in Table 1.

2.1.2. Method for the consolidation experiments

Single plies were consolidated at a pressure of 1 bar and the recommended processing temperature of 385 °C with the setup from Fig. 1, following the consolidation cycle as shown in Fig. 2. Release agents were applied conforming to the manufacturers recommendations. First, a set of baseline experiments was performed, followed by experiments where one of the investigated processing parameters was varied. The full list of experiments is shown in Table 2. The first experiment is performed using the baseline conditions. The second up to the fifth experiment investigated the effect of laminate size; experiment six and seven involved a variation of the tool-ply friction by varying the used release media. Experiment eight to ten were designed to study the role of tool CTE, while the last experiment considered a combination of release media and CTE.

Each experiment was performed three times while nine patches of wavy fibers were evenly selected for each consolidated ply. Surface micrographs were taken for these patches using a Leica M125 microscope at a magnification of 25x. The micrographs were processed with a fiber tracing algorithm [17] to determine the wavelength \( \lambda \) and maximum misalignment angle \( \theta \) of the wavy fibers in each patch. Averaged wave parameters \( (\lambda, \theta) \) were determined by averaging all 27 selected wavy patches from three laminates in each experiment. Previous experiments showed that a waviness-free region exists at the edges of a processed laminate [8,18]. Therefore, the length of the waviness-free edge \( L_w \) was determined for each experiment as well.

2.2. Tool-ply friction measurements

2.2.1. Experimental setup

The friction testing setup from Fig. 3 was used to determine the tool-ply CoF at processing conditions for different combinations of release
This setup operates inside a universal testing machine at the University of Twente. Two temperature-controlled steel pressure blocks apply a normal force and heat to a specimen, which consists of two tool plates treated with release media and a composite ply. The CoF between the tool and the ply can be determined by pulling-out the composite ply from between the tool plates and comparing the tensile force with the applied normal force. The reader is referred to the original description of this setup for more details [11].

### Table 1

Details about the parts used in the single-ply consolidation setup as shown in Fig. 1.

| Component            | Details                                      |
|----------------------|----------------------------------------------|
| Vacuum breather      | Airtech LT800                                |
| Composite            | C/PEEK 0.15 mm UD ply, 59%Vf                |
| Release media        | Varied: listed in Table 2                   |
| Tool plate           | Varied: listed in Tables 2 & 4              |
| Graphite plate       | RX® Egraflex NR, 1 mm                        |
| Vacuum film          | Airtech Thermalimide E, 50 μm               |
| Vacuum sealant       | Airtech A800-3G                              |
| Base plate           | Steel S420MC, 10 mm                          |

### Table 2

Test matrix for the single-ply consolidation experiments. Bold entries indicate the investigated parameter with respect to the baseline experiment. U = Upilex 25S release film, F = Frekote 700-NC release agent. The * indicates the baseline experiment.

| # | Abbr. | Tool plate material | Release film | Release agent |
|---|-------|---------------------|--------------|---------------|
| 1*| S-U-300| Steel              | U            | -             |
| 2 | S-U-100| Steel              | U            | 100           |
| 3 | S-U-150| Steel              | U            | 150           |
| 4 | S-U-200| Steel              | U            | 200           |
| 5 | S-U-450| Steel              | U            | 450           |
| 6 | S-F-300| Steel              | F            | 300           |
| 7 | S-UF-300| Steel            | U + F        | 300           |
| 8 | A-U-300| Alu                | U            | 300           |
| 9 | K-U-300| Kovar              | U            | 300           |
| 10| G-U-300| Graphite           | U            | 300           |
| 11| A-UF-300| Alu              | U + F        | 300           |

### Table 3

Test matrix for the tool-ply friction measurements.

| #  | Abbr. | Tool plate material | Release film | Release agent |
|----|-------|---------------------|--------------|---------------|
| 1  | N     | Steel               | -            | -             |
| 2  | U     | Steel               | Upilex 25S  | -             |
| 3  | F     | Steel               | -            | Frekote 700-NC|
| 4  | M     | Steel               | Marbocote 227CEE|
| 5  | UF    | Steel               | Upilex 25S  | Frekote 700-NC|
| 6  | G     | Steel               | Graphite     | -             |
| 7  | Ua    | Aluminum            | Upilex 25S  | -             |

2.2.2. Method for the tool-ply friction measurements

The friction testing setup from Fig. 3 was used to determine the CoF between a steel tool plate and a C/PEEK composite ply. Release agents were applied to the tool plates and to both sides of the release film, if present. Five specimens were tested for each variation of release media, listed in Table 3. A typical friction test specimen consisted of a 50 mm wide C/PEEK composite ply placed between two 50 mm wide tool plates with one of the six release medium combinations. Additionally, the seventh experiment investigated the effect of tool material. The tool plates were fixed to the surrounding frame and the top end of the composite ply was fixed in the grips of the universal testing machine. The bottom end of the composite ply could move freely between the tool plates. Release films, if used, could move freely between the composite ply and the tool plates. The temperature-controlled blocks were heated to 385°C while the specimen was clamped between the blocks. A normal force \( F_n \) of 250 N was applied to the 50 × 50 mm\(^2\) block surfaces, generating a pressure of 1 bar. A 5 min dwell time was applied to thoroughly heat the specimen. Subsequently, the composite ply was pulled out at a rate of 25 mm/min in the fiber direction. It should be mentioned that the slip rates observed during consolidation are typically one or two orders of magnitude smaller. More realistic slip rates, however, cannot easily be attained with the setup used in this work.

![Fig. 1. Schematic representation of the single-ply consolidation setup.](image1)

![Fig. 2. Temperature profile during consolidation. (1) Application of vacuum to generate a pressure of 1 bar. (2) Heating to \( T_{max} \) (385 °C) at a rate of 5 °C/min. (3) Dwell time of 20 min at \( T_{max} \). (4) Cooling to room temperature at a rate of 5 °C/min. (5) Release of the vacuum and demoulding.](image2)

![Fig. 3. Schematic representation of the friction testing setup, as described in [11].](image3)
Therefore, the assumption is made here that the influence of release media on the friction coefficient at 25 mm/min is representative for lower rates as well. The pulling force $F_t$ was registered with a 1 kN force cell during the experiment. The results were expressed in terms of the CoF between the tool and the ply, as it is the traditional method of describing friction results [13]. The CoF is based on ASTM standard D1894, using simple Amontons-Coulomb friction [19]. The following equation was used to calculate the CoF $\mu$:

$$\mu = \frac{F_t}{2F_n}$$  \hspace{1cm} (1)

with $F_t$ and $F_n$ as shown in Fig. 3. The factor 2 in the denominator corresponds with the sliding surfaces both sides of the composite ply.

2.3. Materials

2.3.1. Composite

The composite material used in the experiments was the Cetex® TC1200 C/PEEK unidirectional prepreg, manufactured by Toray Advanced Composites. A typical ply from this material has a thickness of 0.15 mm, a fiber volume fraction of 59%, a glass transition temperature $T_g$ of 143 °C and a melting point $T_m$ of 343 °C. [20]. The AS4 fibers in the prepreg have a CTE of $-0.63 \times 10^{-6}$ K$^{-1}$ (21). This CTE is assumed to remain fairly constant over the entire processing temperature range, based on CTE measurement for similar types of carbon fibers [22]. All plies and friction test specimens were cut to size from the same 12" wide roll using an ideal 1038 cutter.

2.3.2. Tool material

Tool plates of four different materials were used in the consolidation experiments. Although their CTEs at room temperature are readily available in the literature, their CTEs in the waviness formation temperature interval $\Delta T_w$ are not, except for Kovar. These values are necessary for a fair comparison of the materials, as waviness only forms in the $\Delta T_w$ interval during consolidation [8]. $\Delta T_w$ is defined as the interval between $T_{\text{max}}$ and $T_{\text{cryst}}$, with typical values for C/PEEK of 385 °C and 285 °C respectively. The missing CTEs in the $\Delta T_w$ range could be determined with averaged thermal expansion data from the literature using Eq. (2), where $\Delta T_w$ was rounded to 400–300 °C to match with the literature values:

$$\alpha_{\text{ave}} = \frac{\Delta T_w}{T_{\text{ave}}} = \alpha_{20-400} \Delta T_{20-400} - \alpha_{30-300} \Delta T_{30-300}$$  \hspace{1cm} (2)

with $\alpha$ representing the average CTE and the subscript indicating the temperature range. The calculated CTEs and the references can be found in Table 4.

2.3.3. Release media

One type of release film and two release agents were considered in this research. The release film was an Upilex 25S polyimide film with a thickness of 25 µm. The release agents were Henkel Frekote® 700-NC and Marbocote® 227CEE.

### Table 4

| Tool material | Type | CTE (RT) | CTE ($\Delta T_w$) | Ref. |
|---------------|------|----------|--------------------|------|
| Aluminum      |      | 22       | 28.7               | [23] |
| Steel         | AISI 430 | 11       | 12.6               | [24] |
| Kovar         |       | 5.6      | 4.6 – 5.2          | [25] |
| Graphite      | RX® Egraflex NR | 2.0 | 3.7               | [26] |

3. Results

3.1. Single-ply consolidation

Six out of the eleven sets of consolidation experiments yielded patches of wavy fibers scattered throughout the ply, all in a similar way. A typical result is shown in Fig. 4, along with a schematic representation. No waviness was observed near the edges of the ply perpendicular to the fibers. However, the wavy patches did extend all the way to the edges in the direction transverse to the fibers. The measured waviness parameters and waviness-free edge lengths are shown in Table 5 and Fig. 5. Representative surface micrographs of wavy fiber patches, taken from the plies that did show fiber waviness, are shown in Fig. 6.

3.2. Tool-ply friction

A typical graph of the measured force during a friction measurement is shown in Fig. 7. Initially, the force builds up until it overshoots, followed by a plateau. Another tool-ply friction experiments showed a decreasing overshoot with a decreasing pull-out rate [11]. Therefore, the plateau values for each friction experiment were used to calculate the CoF with Eq. (3), since the tool deformation rate in the consolidation process is considered very low. The resulting CoFs are shown in Fig. 8.

The slip plane alternated between the composite and the release film, and between the release film and the tool when only a release film was used as the release medium (U and $U^\circ$). The experiments in which only release agents were used (F and M) exhibited a slip plane between the composite and the tool, while the experiments where the release film was combined with release agent (UF) showed a slip plane between the release film and the tool. The latter slip plane was observed as well for the experiments where a graphite sheet was used as a release medium (G).

4. Discussion

4.1. Tool-ply friction experiments

A typical curve for the pull-out force during a tool-ply friction measurement is shown in Fig. 7. The standard deviation in these measurements is rather large, and may be explained by thickness variations and inhomogeneities that are present throughout the composite ply [27]. The influence of the various release media on the CoF is shown in Fig. 8. The experimental results show a high tool-ply CoF ($\mu = 0.53$) between untreated steel and a C/PEEK ply. The use of Upilex release film lowered the CoF with approximately 20% ($\mu = 0.42$) compared with untreated steel. No significant difference was observed for Frekote 700-NC ($\mu = 0.26$) or Marbocote 227CEE release agent ($\mu = 0.29$); both decreased the CoF with approximately 50% when directly applied on steel. These results compare well with results for low-velocity friction experiments of C/PEEK in forming conditions [13]. Interestingly, a combination of both release film and release agent decreased the CoF with approximately 95% ($\mu = 0.03$). An explanation for this drastic decrease in CoF may be an enhanced slip layer between the release film and steel tool, since the release agent was applied at this interface as well. The use of a graphite sheet between the steel and the composite lowered the CoF with roughly 90% ($\mu = 0.05$). All tool-ply friction experiments were performed in isothermal conditions with a pull-out velocity of 25 mm/min. However, the formation of waviness is non-isothermal [8] and the tool shrinks with a velocity in the order of 0.01 mm/min during consolidation. Furthermore, the literature shows a dependency of the CoF on the temperature and the velocity [13]. Therefore, the measurements do not provide quantitative values for the COFs during consolidation, however they do provide a valuable comparison between different combinations of release media at processing conditions.
4.2. Influence of release media

The consolidation experiments S-U-300, S-F-300 and S-UF-300 show the influence of different release media on the formed waviness. No significant difference is observed in the wavelength $\lambda$ and the misalignment angle $\theta$ between the baseline (S-U-300) and the experiment where the tool was treated with Frekote 700-NC release agent (S-F-300). However, the waviness-free edge length $L_e$ was significantly larger in the latter experiment. The experiments where Frekote 700-NC was combined with Upilex 25S release film (S-UF-300 and A-UF-300) yielded no fiber waviness.

The increased waviness-free edge length between S-U-300 and S-F-300 can be explained by the decrease in tool-ply friction, as shown in Fig. 8. The release media used in S-U-300 yielded a CoF of 0.42 while the release media combination of S-F-300 resulted in a CoF of 0.26. Analogous to the shear lag theory [28], it is assumed that an axial force on the fiber builds up from the edge of the laminate towards the middle, driven by shear force transfer from the shrinking tool to the ply [18]. This force transfer is governed by tool-ply friction. Therefore, it was expected that a decrease in CoF would lead to an increase in the waviness-free edge length. This is indeed observed between S-U-300 ($\mu = 0.42, L_e = 53$ mm) and S-F-300 ($\mu = 0.26, L_e = 87$ mm). A schematic representation of a ply with a shrinking tool plate on one side is shown in Fig. 9 to further explain this phenomenon.

The shear stress $\tau$ is dictated by the difference in shrinkage between the tool plate and the ply, combined with tool-ply friction. In turn, the tool-ply friction is governed by the pressure $p$ and the CoF $\mu$. The fibers are assumed to be stress-free at the edge of the laminate, while an axial fiber stress $\sigma$ is built up from the edge towards the center. Therefore, the following relation can be established:

$$\sigma(x) = \frac{1}{h} \int_0^L 2\tau \, dx \approx \frac{2}{h} \int_0^L \mu p \, dx,$$

(3)

assuming a constant shear stress, limited by tool-ply slip which is dictated by the pressure and the CoF. The factor 2 represents symmetry: a tool-ply interface is present on both sides of the ply. This equation reduces to Eq. (4), under the assumption that a fiber buckles at the waviness-free edge length $L_e$, where the critical axial stress $\sigma_{crit}$ is reached:

$$\sigma_{crit} = \frac{2}{h} \int_0^{L_e} \mu p \, dx \approx \frac{2\mu L_e}{h}.$$

(4)

Table 5

Test matrix and wave parameters for the experiments. $\# =$ Experiment nr, $U =$ Upilex 25S release film, $F =$ Frekote NC700 release agent, $\lambda =$ wavelength, $\theta =$ maximum misalignment angle. $\bar{z}$ and $\bar{\theta}$ are averaged values for all wavy patches in the corresponding experiment. $L_e$ is the waviness-free edge length. Round brackets indicate the standard deviation. A dash indicates no observable waviness.

| $\#$ | Tool | Release | $L$ | $\lambda$ | $\theta$ | $\bar{z}$ | $\bar{\theta}$ | $L_e$ |
|-----|------|---------|-----|---------|--------|--------|--------|-----|
| 1*  | Steel | U       | 300 | 2.8 (0.3) | 5.3 (1.0) | 2.7 (0.3) | 5.5 (1.1) | 53 (10) |
| 2   | Steel | U       | 100 | 2.9 (0.3) | 5.8 (1.3) | 2.7 (0.3) | 5.5 (1.2) | 51 (11) |
| 3   | Steel | U       | 150 | 2.8 (0.4) | 3.9 (1.1) | 2.6 (0.3) | 4.6 (1.1) | 52 (10) |
| 4   | Steel | U       | 200 | 2.7 (0.2) | 5.0 (0.8) | 2.6 (0.3) | 4.9 (1.1) | 51 (9) |
| 5   | Steel | U       | 450 | 2.7 (0.4) | 5.2 (0.9) | 2.5 (0.3) | 4.9 (1.2) | 51 (9) |
| 6   | Steel | F       | 300 | 2.8 (0.2) | 5.1 (1.1) | 2.7 (0.3) | 5.6 (0.7) | 52 (10) |
| 7   | Steel | U + F   | 300 | 2.6 (0.4) | 6.1 (1.2) | 2.5 (0.3) | 5.9 (1.3) | 52 (10) |
| 8   | Alu   | U       | 300 | 4.2 (0.6) | 15.4 (2.7) | 4.1 (0.5) | 15.1 (2.6) | 43 (13) |
| 9   | Kovar | U       | 300 | 4.2 (0.6) | 15.7 (2.6) | 4.2 (0.4) | 14.1 (2.1) | 44 (14) |
| 10  | Graphite | U       | 300 | 4.0 (0.4) | 14.3 (2.1) | 3.9 (0.3) | 14.2 (2.0) | 45 (15) |
| 11  | Alu   | U + F   | 300 | 4.0 (0.4) | 14.3 (2.1) | -       | -       | -    |

* Baseline experiment.

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$$\sigma_{crit} = \frac{2}{h} \int_0^{L_e} \mu p \, dx \approx \frac{2\mu L_e}{h}.$$

(4)
The critical buckling stress is assumed to be constant for the applied lay-up, material and pressure. These parameters did not vary between all consolidation experiments. Consequently, the product $\mu L_e$ should be constant as well for these experiments. This is indeed the case, despite the significant difference in CoF and waviness-free edge length between S-U-300 and S-F-300: for S-U-300 the product is 22.3 mm and for S-F-300 the product is 22.6 mm. Even with aluminum tool plates (A-U-300) the product is close to these values: 21.07 mm. If the value of 22.3 mm is used as a reference value, it would mean that the waviness-free edge length for a low-CTE experiment such as S-UF-300 becomes very large: $22.3/0.03 = 743$ mm. This may explain the absence of waviness in the experiments with a very low CoF.

The presence of a waviness-free edge length has implications for the critical part length. As a logical consequence, parts that are smaller than twice the waviness-free edge length should be free of fiber waviness. The waviness-free edge length of around 52 mm for the S-U experiments implies that a laminate with a length of 104 mm should be free of waviness. This may explain the absence of fiber waviness in experiment S-U-100. A critical laminate length for the formation of in-plane waviness has been observed by other researchers as well, who focused on the autoclave consolidation of C/PSU laminates [6,18]. Additionally, experiments S-U-150 up to S-U-450 indicate that the laminate size does not affect any of the waviness parameters ($\lambda$, $\theta$, $L_e$). Similar observations were made in another study, where the amplitude and wavelength only showed a slight upward trend with the part length, while an increased number of wavy patches was among the most significant observations affected by the part length [18]. Larger parts need larger tool plates, which have a larger tool shrinkage. It is argued that the imposed tool shrinkage is divided over the wavy patches along the length of the part, resulting in a similar amount of tool shrinkage per wavy patch, since the waviness parameters in the patches were not affected by the laminate size.

Fig. 5. Resulting maximum misalignment angle ($\theta$), wavelength ($\lambda$) and waviness-free edge length ($L_e$) for the experiments that yielded fiber waviness. Error bars indicate the standard deviation. (-) represents no observable waviness.

Fig. 6. Surface micrographs of typical wavy fibers.

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The influence of the tool CTE

The relevant CTEs of the tool materials used in this research are listed in Table 4. The CTEs of the steel, aluminum and graphite tool plates increase as the temperature increases. Hence, the values in the waviness formation temperature interval were calculated for these materials for a fair comparison. The CTE of Kovar does remain constant over the entire processing temperature range [25]. Experiments S-U-300, A-U-300, K-U-300 and G-U-300 show a significant influence of tool material on the waviness parameters $\lambda$ and $\theta$: the low-CTE Kovar and graphite tool plates yielded no waviness, while $\lambda$ and $\theta$ increased for an increase in CTE between steel and aluminum tool plates. These results are in agreement with the results for the amorphous C/PSU material system [6,7]. Experiment A-U300 combined a high CTE with a very low CoF. This experiment demonstrated that waviness-free laminates are possible even with high-CTE tool plates. Finally, the use of graphite tool plates (G-U-300) to produce waviness-free laminates has already been suggested in the literature, substantiated by the low CTE of graphite [6]. In addition, the research presented in this paper suggests that the low CoF of graphite alone is enough to produce waviness-free laminates.

4.4. General discussion

The typical distribution of wavy patches on the surface of a consolidated ply, as illustrated in Fig. 4, may be explained by an inhomogeneous distribution of tool-ply CoF over the surface. The large spread in friction values (Figs. 7 and 8) support this hypothesis. It should be mentioned that inhomogeneities in the temperature distribution, pressure distribution and material may also affect the distribution of the wavy patches. The distance in the fiber direction between wavy patches may be explained by locally reduced fiber stresses in a wavy patch; the distance is necessary to build up sufficient fiber stresses for the next wavy patch to form.

The tool-ply CoF was identified as a key parameter in the formation of fiber waviness during laminate consolidation. This study shows that the production of waviness-free laminates is possible even with high CTE tool plates, provided that the tool-ply CoF is sufficiently low or the laminate size is sufficiently small. From the point of view of industrial practice it is advised to focus on a low CoF between the tool and the ply, since the laminate size is usually constrained by the part design. Although this research shows the potential for very low CoFs by combining a release film with a release agent for flat laminates, the use of release media may be limited for more complex shaped parts such as U-shaped laminates. In those cases it would be more effective to use tool plates with a sufficiently low CTE. Moreover, the requirements for the CoF become more stringent with an increasing part size, as indicated by the proposed relation in Eq. (4). Alternative ways to reduce the tool-ply CoF may be to lower the processing temperature, lower the deformation rate, or increase the pressure [13,15]. However, in practice these parameters are constrained by other properties such as the degree of ply bonding, the void content and the processing time. It is worth mentioning that the importance of the CoF rationalizes other research topics focused on tool-ply interactions during part manufacturing, such as laser-induced mold surface treatments [29] or the polymeric interface structure between the tool and the ply [30].

The experiments considered in this research are limited to out-of-autoclave consolidation, as the pressure applied to the laminates did not exceed 1 bar. In addition, flow-induced waviness is minimized by choosing low pressures, making it easier to isolate the effects of tool shrinkage. Although the proposed relation in Eq. (4) indicates that a higher pressure should lead to a smaller waviness-free edge length, a more elaborate experimental study is necessary to see whether practice confirms this relation or that other effects play a role. Despite the low pressure, we believe that the findings in this study do provide an insight into waviness formation during autoclave or press consolidation processes, as the temperature cycles, tooling materials, and lay-ups in those processes are comparable to the experiments presented in this study.

5. Conclusions

The influence of tool material, release media and laminate size on the occurrence of in-plane fiber waviness in C/PEEK plies has been discussed in this paper. A vacuum-assisted oven consolidation setup was used to consolidate single plies between a combination of tool plates and several release media. The degree of waviness was quantified for each of these combinations. Furthermore, a friction testing setup was used to determine typical coefficients of friction (CoF) between the tool material and the composite ply with varying release media.

The experimental results show a high tool-ply CoF ($\mu = 0.53$) between untreated steel and a C/PEEK ply sliding in the fiber direction. The CoF dropped with 20% ($\mu = 0.42$) when Upilex 25S polyimide film was used as the release medium. Tool-ply friction reduced with approximately 50% when Frekote® 700-NC ($\mu = 0.26$) or Marbocote® 227CEE ($\mu = 0.29$) release agent were used. A drastic decrease of approximately 95% is realized when the release film was combined with a release agent ($\mu = 0.03$). A graphite sheet used as a release film yielded a CoF of $\mu = 0.05$.

This research provides additional evidence that tool shrinkage is indeed the driving force for the formation of fiber waviness in C/PEEK laminate consolidation. The experimental results show that the degree of fiber waviness does not depend on the laminate size in the fiber
direction. This holds for laminates larger than twice the waviness-free edge length; no waviness is formed if the laminate is smaller. The wavelength and maximum misalignment angle showed no dependency on the CoF. However, the waviness-free edge length directly depended on the CoF, with an increased edge length for a decreased tool-ply CoF. The wavelength and maximum misalignment angle were influenced by the coefficient of thermal expansion (CTE) of the tool material, provided that the CoF is sufficiently high to form waviness: single C/PEEK plies exhibited no waviness when graphite or Kovar tool plates were used, a moderate wavelength and maximum misalignment angle when steel tool plates were used and a large wavelength and maximum misalignment angle when aluminum tool plates were used.

The formation of in-plane fiber waviness in the consolidation process can, on the one hand, be prevented by choosing a tool material with a low CTE, regardless of the tool-ply CoF. On the other hand, ensuring a low tool-ply CoF will prevent fiber waviness, regardless of the tool material. It can be concluded that the formation of waviness in laminate consolidation depends on the balance between the CTE of the tool, the tool-ply CoF and the laminate size.

Declaration of Competing Interest

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.

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