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Intimate contact development during laser assisted fiber placement: Microstructure and effect of process parameters

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ABSTRACT

Intimate contact development under LAFP-specific thermal and mechanical boundary conditions/interactions and the effect of process parameters are investigated. One-layer, unidirectional strips of CF/PEKK material were placed with different process parameters on a flat tool surface to create different intimate contact conditions. The concept of effective intimate contact, which is based on the resin content at the surface, is introduced and a methodology to measure it from surface micrographs is provided. Degree of effective intimate contact measured from the samples was compared with the existing intimate contact models. The temperature history in the compaction zone was estimated with a finite element model and pressure sensitive films were used to determine the compaction pressure. It is shown that in addition to the squeeze flow mechanism, which is the base for the current intimate contact models, through-thickness percolation flow of the resin needs to be considered to explain the effective intimate contact development.

1. Introduction

Thermoplastic composites (TPC) have drawn significant interest in the aerospace industry for their infinite shelf life, high fracture toughness, chemical and solvent resistance, weldability and recyclability [1–3]. Automated fiber placement (AFP) has become a promising manufacturing solution for TPC, providing reduction in labor time per component, lower material scrap rates and higher repeatability [4]. In modern laser assisted fiber placement (LAFP) systems, a laminate is built by heating the surface of the tape and substrate with a laser heat source pointed towards the nip point and compacting with a flexible roller as shown in Fig. 1.

Due to the re-melting capability of thermoplastic polymers, in-situ consolidated (i.e., without a post-consolidation step in an autoclave, oven or press) structures can be manufactured with the LAFP process. This aspect of the technology is especially appealing since significant capital investment and operating costs related to post-consolidation can be saved if sufficient part quality with a feasible processing speed is achieved. In order to achieve this goal, extensive research has been conducted in the last decades to understand the mechanisms which play a role at the heating, compaction and cooling phases of the process [5–14].

The compaction phase is especially critical since two subsequent plies consolidate only with the pressure supplied by the compaction roller. Intimate contact development is the first step of consolidation and affects several aspects during the process which significantly influence the properties of the final structure. Firstly, bond strength formation between subsequent plies requires intimate contact development between surfaces. Bonding is driven by diffusion and entanglement of polymer molecules across the interface through reptation [15]. This mechanism is active only at the locations which are in intimate contact [16]. Secondly, inter-laminar void content in the final structure is a direct result of intimate contact development. Such voids act as crack initiation points and reduce the mechanical performance of the final product, especially inter-laminar shear, compression and transverse tensile strength and fatigue life [17]. Finally, heat transfer might be reduced at the ply interfaces due to incomplete intimate contact [10,18]. The effect of reduced heat transfer on the temperature history influences all temperature related phenomena during LAFP, e.g. crystallinity, healing, void compaction/decompaction and residual stresses. Understanding how intimate contact develops during LAFP is therefore an important step for linking the process parameters to the final quality of the parts.

The common approach to explain how intimate contact develops during thermoplastic composite processing has been based on the squeeze flow of the surface asperities. Such models combine the compaction force, an idealized description of the composite surface and the temperature-dependent homogenized viscosity of the composite...
material to find the degree of intimate contact given the temperature and pressure history of the process. Using this approach, several models were proposed for intimate contact development. The first model was proposed by Dara and Loos for isobaric and isothermal conditions [19]. The squeeze flow mechanism was justified by the high viscosity of the thermoplastic polymers at melting temperature. They modeled the composite surface as a collection of rectangular elements with non-uniform height and width, approximated by a 2-parameter Weibull distribution. Lee and Springer [20] followed a similar rationale but simplified the surface representation with uniform rectangles. Mantell and Springer [21] extended this model for non-uniform pressures and temperatures, which is more suitable for automated fiber placement or winding. Yang and Pitchumani suggested that the asperities in the thermoplastic composite surfaces exhibit fractal characteristics and proposed a surface representation with a fractal Cantor set [22].

These models have been extensively used to predict intimate contact development for the AFP process [23–26]. However, limited prediction accuracy was reported for modern systems with laser heaters [26–28]. This raises the question of whether the squeeze flow mechanism with homogenized material assumption is completely suitable to explain intimate contact development during LAFP. Rapid heating to high processing temperatures (370–400 °C for carbon fiber reinforced polyetherketonketone (CF/PEKK) [29] and carbon fiber reinforced polyetheretherketone (CF/PEEK) [30] tapes) without pressure application and very short compaction times (usually between 0.01 and 0.3 s depending on the deformation of the compaction roller and the placement speed) are distinctive features of LAFP. Moreover, heating above the melting temperature of the resin material without pressure application results in a fiber-rich and rough surface until the tape reaches the nip point [31]. Considering that a significant portion of the tapes already has a resin-poor surface in their original state such as the one shown in Fig. 2 and the microstructure of the tape changes during the heating phase, intimate contact development might not be adequately described with the squeeze flow mechanism. Intimate contact models mentioned above were validated by press-consolidated samples with compaction times orders of magnitude higher than typical compaction times for LAFP, which might suppress the effect of non-uniform microstructure at the surface. Considering the state-of-the-art literature, how intimate contact develops at short time spans typically found in the LAFP process, taking the microstructural changes due to laser heating into account, is still an open question.

Another reason for the lack of knowledge about the mechanisms involved in intimate contact development is how the degree of intimate contact is measured. The degree of intimate contact was measured mostly at the interfaces between the plies [19,22,23,32], although the available intimate contact models consider flattening of a single composite surface against a rigid surface. There is only one study where the predicted intimate contact was compared with the surface of press-consolidated individual tapes [20]; however, the details of how the degree of intimate contact was measured was omitted. Moreover, the final state of the composite interface contains little information about the process of intimate contact development during compaction. Therefore, a more detailed analysis of the intimate contact development on a single tape under LAFP conditions is required.

The aim of this paper is therefore to understand the intimate contact development mechanism under LAFP-specific thermal and mechanical boundary conditions/interactions. Specifically, the following questions will be addressed: How can the degree of intimate contact be quantified considering the inhomogeneity of the composite surface? What is the effect of LAFP process parameters on intimate contact development? Can the squeeze flow mechanism fully explain the intimate contact development during LAFP?

To answer these questions, one-layer samples were prepared with a LAFP machine with different process parameters. The final degree of
intimate contact of the samples was measured with a novel method which considers the non-homogeneous nature of the contact surface. Experimental findings were compared to and contrasted with the state-of-the-art intimate contact models. In order to do that, a heat transfer model of the process was formulated to estimate the temperature history of the tape in the compaction region. The estimated temperature history was then combined with the measured roller pressure and the degree of intimate contact was calculated. The results were used to present a new approach to describe the intimate contact development during LAFP.

2. Materials and methods

2.1. Fiber placement system

Fiber placement experiments were conducted at Royal Netherlands Aerospace Center (NLR). A six-axis articulated robot on a linear axis provided by Coriolis was used. The machine was able to deliver eight 6.35 mm (1/4 in.)-wide tapes simultaneously up to 800 mm/s placement speed. The end effector was equipped with a 6 kW Laserline LDF series diode laser system and an optic lens which created a 56 mm × 28 mm rectangular illuminated area at the 250 mm focal distance. A conformable compaction roller with 60 shore hardness and a diameter of 70 mm was installed on the machine. Fig. 3 depicts the fiber placement system used for this study.

2.2. Specimens for intimate contact investigation

The material used in this study was TC1320-1 CF/PEKK supplied by TenCate, in 1/4 in. slit tapes. Eight tapes with cross-sectional dimensions 6.35 mm × 0.15 mm were placed next to each other simultaneously along 1000 mm long strips on a flat aluminum tool with each combination of process parameters shown in Table 1. The machine parameters were chosen such that they cover the range between the minimum and maximum compaction forces and placement speeds relevant to the industrial use of LAFP at processing temperatures suggested by the manufacturer [29]. The temperature of the heated tool was set to 155 °C to allow the compacted tape to cool down to below its glass transition temperature (159 °C [29]), so that the amorphous phase of the PEKK polymer stays in the glassy state and does not cause further changes in the microstructure after compaction. The tool surface was covered with 50 μm thick polyimide (thermimide) film to reduce the possible effects of the scratches on the tool on the degree of intimate contact measurements. Samples were cut from the mid-section (between 400 and 600 mm from the beginning) of the strips so that the acceleration and deceleration phases of the placement head did not affect the microstructure.

2.3. Process temperature measurement

The surface temperature of the visible nip point of the tape was measured using a long wave infrared (LWIR) camera. The apparent emissivity of the material is required for measuring the temperature with this method. To determine the apparent emissivity, a single layer of material was placed on the tool surface and a thermocouple was attached on the material surface. The material was heated by heating the tool and sufficient time was given to reach the steady state in the material. The end effector of the tape placement robot was positioned such that the thermal image was similar to actual placement conditions. Thermocouple readings were compared with thermal image readings of the same location. The calibration was performed around 220 °C and the apparent emissivity was found to be 0.85 for the CF/PEKK material.

2.4. Pressure measurement

Although the process is controlled by the so called “compaction force”, the actual pressure distribution under the compaction roller is unknown. During placement, the compaction roller applies the pressure while rotating. However, the effect of roller rotation on the normal pressure is negligible and the dynamic case can be simplified into a static one [33]. Fujifilm Prescale LLW pressure sensitive films were used to measure the contact area under the compaction roller. Pieces of the pressure sensitive film were fixed on the tool and different compaction forces were applied with the placement robot statically, as demonstrated in Fig. 4. A force range of 100 to 800 N was covered with steps of 100 N. Then, the dimensions of the stains on the pressure films were measured. It was assumed that the pressure distribution was uniform and the applied force was divided by the measured area to calculate the average compaction pressure.

3. Effective intimate contact

The resin-rich area on the surface is a pre-requisite for bond strength development between the subsequent layers, which is explained with the autohesion theory [7]. Therefore, the degree of intimate contact is meaningful only if it consists of resin-rich areas. However, current intimate contact models assume that the surface asperities are homogeneous. Considering the fiber-rich nature of the tape surface prior to compaction, there is a need for a new definition of the intimate contact phenomenon. Within a region of interest, the ratio of the compacted resin-rich area to the whole projected area is designated as the degree of effective intimate contact (DEIC) in this work.

3.1. Characterization

The methodology to obtain the degree of effective intimate contact is summarized in Fig. 5. Micrographs were taken from the tool side of the surfaces with a Keyence VHX-2000 system equipped with a VH-
Z100 lens. Five images were taken for each specimen with 200× magnification (image size approximately 1.6 × 1.2 mm²). In the first step, grayscale intensity histograms were generated for each image using the software of the microscope. These histograms show how many pixels are present for each grayscale value in an interval of 0 and 255 where 0 is black and 255 is white. Peaks in such a histogram designates the clusters of pixels that belong to certain features in the image \[34\].

Two classes of histograms were observed. The first class of histograms belonged to the samples which have significant amount of resin-rich area on the surface after compaction. There were two clear peaks which represent the dry fibers (the peak with a lower greyscale value) and the compacted resin area (the peak with a higher greyscale value). For each image, a threshold was determined at the local minimum between the two peaks. The pixels with a grayscale value higher than this threshold were selected as the area in effective intimate contact. The second class of histograms were observed for the samples with a low amount of resin on the surface after compaction. Only a single peak was present in these histograms, making it difficult to set a threshold value. In such cases, the threshold was determined based on the point where the slope of the histogram curve is the closest to zero after the first peak as the grayscale intensity value increases. For both cases, this selected area included a low amount of dry fibers in addition to the compacted resin-rich areas.

In an attempt to eliminate the dry fibers from the selected area, all sub-areas smaller than 500 μm² were excluded. This value was found to be appropriate for selecting the resin-rich areas as a result of trials with different minimum area thresholds on many images.

Two reference cases, unprocessed and fully compacted tapes, were examined with the proposed methodology to demonstrate its validity on the extreme cases and compare with the available intimate contact models. Fully compacted samples were created by compacting a single layer of a 30 mm × 30 mm of the same material in a press at a temperature of 380 °C and a pressure of 2 MPa for 20 min.

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**Fig. 4.** (a) Static pressure application with the LAFP machine (b) a stained pressure film with the assumed uniform pressure area. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

**Fig. 5.** Measurement of degree of effective intimate contact from histograms of optical microscope images. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
parameters were determined based on prior experience with CF/PEKK materials. Microstructure of the LAFP-processed strips were qualitatively investigated with cross-sectional microscopy for more insight about effective intimate contact development. Samples were embedded in epoxy and polished for this purpose. The same microscopy system is used to obtain the images.

3.2. ANOVA analysis

The analysis of variance (ANOVA) method was used to evaluate the effects of the process parameters given in Table 1 on the mean of the degree of effective intimate contact measurements. Matlab function anovan was used for this purpose [35]. The outputs of this function were p-values for each process parameter. p-values smaller than 0.05 imply that the mean response of the specific parameter is different from the mean of all data within a confidence interval of 95%. The effects of such a parameter can be considered statistically significant.

3.3. Intimate contact calculation based on squeeze flow models

Effective intimate contact measurements were compared with the squeeze flow-based intimate contact models. As explained in the following section, the temperature and pressure history of the material are required to calculate the final degree of intimate contact. The temperature history of the tape during compaction was calculated with a heat transfer model. The compaction pressure during the process was determined using the methodology presented in Section 2.4. These two process-induced parameters were then fed into two intimate contact models available in the literature.

3.3.1. Intimate contact models

Two squeeze flow-based intimate contact models were used to predict intimate contact development on a single tape processed with LAFP. The first model proposed by Lee and Springer [20] approximates the surface of the tape as a series of uniform rectangles. The degree of intimate contact can be found with the following equation:

\[
D_{ic}(t) = \frac{1}{1 + \frac{P_{app}}{P_{cmp}}} \left[ 1 + 5 \left( 1 + \frac{n_0}{b_0} \right) \frac{a v}{b v} \int_{0}^{t} \frac{P_{app}}{\mu(T)} dt \right]^5
\]

(1)

where \(n_0\) and \(a v\) are geometrical parameters defining the uniform rectangular representation, \(T\) is the temperature, \(P_{app}\) is the compaction pressure, \(t\) is the compaction time and \(\mu(T)\) is the temperature dependent viscoelasticity of the material. The geometrical parameters were obtained from real surface measurements with a procedure similar to the work of Schaefer et al. [32]. Surface profiles were measured with an Olympus LEXT OLS3100 confocal laser scanning microscope. The parameters \(n_0\) and \(a v\) were found by calculating the average of the ratio of the material and gaps at different height levels within \(\pm 2\sigma\) from the mean surface height, where \(\sigma\) designates the standard deviation.

The second model proposed by Yang and Pitchumani [22] approximates the composite surface as a fractal Cantor set and calculates the degree of intimate contact as follows:

\[
D_{ic}(t) = \frac{1}{f} \left[ \frac{\left( \frac{b_v}{b_0} \right)^2 \frac{2^{n+4} + 6^{n+1} + 1}{(n+1)^2} \int_{t_{(n+1)}}^{t}(\frac{P_{app}}{\mu(T)}) dt + 1}{f/t_{(n+1)}} \right]^{\frac{1}{2}}
\]

(2)

for \(t_{(n+1)} \leq t \leq t_{(n)}\)

where \(f\) is the fractal scaling factor, \(n\) is the generation number of the Cantor set, \(D\) is the fractal dimension and \(\frac{n_0}{b_0}\) is the ratio of the height of the first generation asperities to the Cantor block length. These parameters were determined as described in the original paper from real surface profiles and their power spectra. The rest of the parameters are the same with Eq. (1).

Fig. 6 demonstrates the corresponding idealized surface

![Fig. 6. Corresponding idealized surface representations on a real surface profile. (a) Uniform rectangle approximation and (b) Cantor set approximation (only first three generations are shown for clarity). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)](image)

| Table 2 | Surface parameters used in the intimate contact models. |
|---|---|---|---|
| Surface model | Parameter | Explanation | Value |
| Lee and Springer [20] | \(n_0/b_0\) | Ratio of asperity width to gap width | 1.11 |
| | \(a v/b_0\) | Ratio of asperity height to asperity width | 0.57 |
| Yang and Pitchumani [22] | \(f\) | Scaling factor | 1.09 |
| | \(D\) | Fractal dimension | 1.63 |
| | \(n_0/L_0\) | Ratio of first generation asperity height to Cantor block length | 0.20 |
| | \(n\) | Total number of generations | 15 |
representations for both intimate contact models on an example measured surface profile. The average geometrical parameters used for the intimate contact models are summarized in Table 2.

$T$ was calculated from the heat transfer model in Section 3.3.2. $P_{app}$ was obtained from static contact measurements described in Section 2.4. $t$ was calculated by dividing the contact length obtained from the pressure sensitive films by the placement speed. $\mu(T)$ for CF/PEKK material was adapted from [36]. It is expressed as the following relation:

$$\mu(T) = 4.362 \times 10^7 e^{-0.0401(T-623)} \text{Pa s}$$ (3)

### 3.3.2. Heat transfer model

A three dimensional Lagrangian finite element (FE) model of the placed tape and the tool with a moving placement head configuration, based on authors’ previous work [37], was used to estimate the transient through-thickness temperature distribution during compaction. Abaqus 2017 finite element package was used to create and solve the model.

The model solves the energy equation to find the transient temperature distribution $T(t)$ within the bodies, assuming the internal heat generation is negligible:

$$\rho C_p \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T)$$ (4)

where $\rho$ is the density, $C_p$ is the specific heat and $k$ is the conductivity of the material. Referring to Fig. 7, the following boundary conditions were applied:

The aluminum tool was assumed to have a fixed temperature $T_{tool}$ due to its large thermal mass. The incoming tape was assumed to reach uniform through-thickness temperature $T_{tape}(0)$ at the visible nip point. Past the visible nip point, the tape cools down due to the contact with the compaction roller in the shadow and compaction zones (see Fig. 1). Following that, the ambient air cools down the tape at the top surface. The heat loss to the compaction roller and the ambient air after compaction was set to a very low value, namely $10^{-5}$ W/m² K. The thermal contact conductance is active were updated with an Abaqus FILM subroutine. At the regions in front of the nip point, no heat transfer was possible between the tape and the tool. To impose this condition without creating numerical problems, the thermal contact conductance was set to a very low value, namely $10^{-5}$ W/m² K. The thermal contact conductance was set to $10^6$ W/m² K past the nip point, assuming almost perfect heat transfer occurs at the interface between the tape and the tool.

The temperature history of the tape was calculated for the scenario where the highest intimate contact is expected (the lowest placement speed $(v)$ and the highest visible nip point temperature $(T_{tape}(0))$, tool temperature $(T_{tool})$ and compaction force in Table 1). The roller contact length at the compaction zone, $L_c$, was determined from the contact length measurements described in Section 2.4. The contact length for a compaction force of 800 N was used since it was observed that this parameter did not affect the temperature history significantly. The length of the shadow, $L_s$, was estimated from the computer aided design (CAD) drawings of the end effector. The process related and geometric parameters used in the model are summarized in Table 3. Thermal material properties provided in Table 4 were used for the composite tape.

DC3D8 linear hexahedral heat transfer elements were used to mesh the tape and the tool. The tape was meshed with 10, 120 and 6 uniform elements in the thickness, length and width directions, respectively.

### 4. Results

#### 4.1. Pressure measurement

The average compaction pressure has a nonlinear relationship with the applied compaction force, as seen in Fig. 8. The compaction roller deforms more as the compaction force increases, creating a larger contact area. Even though a larger contact area is favorable for better intimate contact development, it poses a limitation on reaching high pressure. Comparison of the applied compaction force and average pressure under the roller provides useful information for further discussions.

#### 4.2. Surface and cross-sectional micrographs

Effective intimate contact measurements of unprocessed tapes and fully consolidated samples are summarized in Table 5 along with the predictions made with Eqs. (1) and (2). Reflecting the fiber-rich structure of the surface, the degree of effective intimate contact was very low for the unprocessed tow. On the contrary, press consolidated samples

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Fig. 7. Modeling space and moving boundary conditions in the FE model. $h$ and $C$ represent the applied convection coefficient and thermal conductance, respectively. For the other parameters, please refer to Table 3. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)
showed almost complete effective intimate contact. The major difference between the predictions and measurements can be observed for the unprocessed tape. Both squeeze flow models indicate that the unprocessed tape is in considerable intimate contact with a hypothetical opposite surface, as demonstrated in Fig. 6. However, since the models consider only the surface profiles and not the microstructure, the effect of resin content at the surface is not accounted for. The effective intimate contact concept illustrated in Fig. 5 provides a methodology which is sensitive to the resin content at the surface and able to measure the whole range of degree of effective intimate contact (i.e., 0–100%)

for the fiber-placed samples.

The surface micrographs shown in Fig. 9 demonstrate the morphology of the contact surface for a range of effective intimate contact levels. In Fig. 9a, a specimen with a low degree of effective intimate contact (1%) is shown. The surface exhibits very low amounts of resin content and the surface is covered with dry fibers. In Fig. 9b, a surface with slightly higher amount of degree of intimate contact (23%) is shown. Squeezed out and flattened resin portions are apparent. Fibers are covered with this squeezed-out resin locally. Flattened resin regions mostly elongate along fibers since dry fibers restrict the motion of the molten resin more in the perpendicular direction. The micrograph in Fig. 9c shows a high degree of effective intimate contact obtained within this study (57%), where more resin is squeezed out and covers a larger portion of the surface. The constraining effect of the dry fibers can still be observed on the wetting behavior of the resin material. Within large impregnated areas, some gaps elongated in the fiber direction exist.

Representative cross-sectional micrographs of specimens manufactured with the same nip point temperature (400 °C) and placement speed (100 mm/s) but different compaction forces (100N and 800N) are shown in Fig. 10. Such a comparison is informative about the change in the microstructure during the intimate contact development. Fig. 10a shows a sample with low intimate contact development (22.2%) due to low compaction force. In addition to flattened fiber-resin mixture, incompletely compacted (non-flat) resin-fiber mixture and loose dry fibers can be observed at the surface on the tool side. The specimen in Fig. 10b represents a specimen with higher degree of intimate contact (54.8%) since higher compaction force results in more deformation at the surface given the same thermal history (Eqs. (1) and (2)). Regions with resin-fiber mixture at the surface on the tool side are mostly flattened. Dry fibers are present; however, it can be observed that they are compressed more than the case with the lower compaction force.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tape temperature prior the visible nip point</td>
<td>$T_{tape}(0)$</td>
<td>400</td>
<td>°C</td>
</tr>
<tr>
<td>Fixed tool temperature</td>
<td>$T_{tool}$</td>
<td>155</td>
<td>°C</td>
</tr>
<tr>
<td>Placement speed</td>
<td>$v$</td>
<td>100</td>
<td>mm/s</td>
</tr>
<tr>
<td>Compaction pressure (for intimate contact models)</td>
<td>$P_{app}$</td>
<td>0.29–0.86</td>
<td>MPa</td>
</tr>
<tr>
<td>Roller contact length in the compaction zone</td>
<td>$l_c$</td>
<td>4</td>
<td>mm</td>
</tr>
<tr>
<td>Roller contact length in the shadow zone</td>
<td>$l_s$</td>
<td>100</td>
<td>mm</td>
</tr>
<tr>
<td>Tape thickness</td>
<td>$t_{tape}$</td>
<td>0.15</td>
<td>mm</td>
</tr>
<tr>
<td>Tool thickness</td>
<td>$t_{tool}$</td>
<td>30</td>
<td>mm</td>
</tr>
<tr>
<td>Tape width</td>
<td>$w_{tape}$</td>
<td>50.8</td>
<td>mm</td>
</tr>
</tbody>
</table>

Table 3

The process related and geometric parameters used in the heat transfer model.

Table 4

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>$\rho$</td>
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<td>kg/m³</td>
</tr>
<tr>
<td>Thermal conductivity (W/m °C)</td>
<td>—</td>
<td>990</td>
<td>50</td>
</tr>
<tr>
<td>—</td>
<td></td>
<td>1176</td>
<td>100</td>
</tr>
<tr>
<td>—</td>
<td></td>
<td>1353</td>
<td>150</td>
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<tr>
<td>—</td>
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<td>1675</td>
<td>200</td>
</tr>
<tr>
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<td></td>
<td>1865</td>
<td>250</td>
</tr>
<tr>
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<td></td>
<td>2220</td>
<td>300</td>
</tr>
<tr>
<td>—</td>
<td></td>
<td>2289</td>
<td>350</td>
</tr>
<tr>
<td>Specific Heat (J/kg °C)</td>
<td>—</td>
<td>6</td>
<td>0.73</td>
</tr>
<tr>
<td>Temperature (°C)</td>
<td>—</td>
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</tr>
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</table>

Table 5

Degree of effective intimate contact of the reference cases and degree of intimate contact predictions with the currently available models.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measured DEIC (%)</td>
<td>0.2 ± 0.4</td>
</tr>
<tr>
<td>Lee &amp; Springer (%)</td>
<td>47.4</td>
</tr>
<tr>
<td>Yang &amp; Pitchumani (%)</td>
<td>27.5</td>
</tr>
</tbody>
</table>

Unprocessed Tape

Press Consolidated Ply (at 380 °C, 20 min) 96.6 ± 1.8 100 100

Fig. 8. Nonlinear relation between the applied compaction force and the average compaction pressure measured with pressure sensitive films due to the increasing contact area.
4.3. Effects of process parameters on intimate contact development

Final degrees of effective intimate contact at different tool temperatures, nip point temperatures, compaction forces and placement speeds are summarized in Fig. 11. ANOVA analysis is applied on this data set to find statistically significant parameters on effective intimate contact development.

4.3.1. ANOVA analysis

The results presented in Table 6 indicate that the sources tool temperature (TT), compaction force (CF), placement speed (PS) and the interaction terms TT*CF and TT*PS caused statistically significant differences on the degree of effective intimate contact within the parameters presented in Table 1. For the other sources, it is not possible to distinguish the effects from experimental scatter. Significance of the interaction terms (sources TT*CF and TT*PS) shows that the effect of the tool temperature dominates over the others. This is also evident in Fig. 11: samples placed on the cold tool (top row) have very low degrees of effective intimate contact compared to the samples placed on the hot tool (bottom row) and effects of compaction force and placement speed are not apparent. Therefore, in order to assess the effects of compaction force and placement speed, the results from the hot tool should be used.

Interestingly, the target nip point temperature (source NPT) did not cause significant changes on the degree of effective intimate contact. Although the squeeze flow viscosity of CF/PEKK decreases more than twice when the temperature increases from 380 °C to 390 °C according to Eq. (3), this decrease was not reflected in the results as an increase in effective intimate contact development. A possible explanation is the very short time spent above the melting temperature due to the quenching effect of the tool. Hence, data from two different nip point temperatures were combined to present the results of the effects of compaction force and placement speed.

4.3.2. Compaction force

The effect of the compaction force on the degree of effective intimate contact is shown in Fig. 12. The horizontal axis of the figure is the compaction force and the vertical axis is the degree of intimate contact. The figure shows that as the compaction force increases, the degree of intimate contact increases as well. The degree of intimate contact predictions based on the calculated temperature history of the top and bottom sides of the tape are shown in Fig. 14. In the non-illuminated shadow zone, the temperature of the tape decreases unevenly in the thickness direction until it reaches the nip point (the beginning of the consolidation zone). The top surface of the tape cools down faster than the bottom side due to direct contact with the roller. When the tape reaches the nip point, the top and bottom surfaces are at 383 °C and 375 °C, respectively. However, as soon as the tape enters the consolidation zone, the bottom surface of the tape quenches to the mold temperature rapidly (Fig. 14b). This causes the bottom surface to stay above the melting temperature for only 2.5 ms. On the other hand, the top surface stays above the melting temperature for 15 ms, a considerably longer duration.

4.4. Temperature history and intimate contact predictions

The calculated temperature history of the top and bottom sides of the tape are shown in Fig. 14. In the non-illuminated shadow zone, the temperature of the tape decreases unevenly in the thickness direction until it reaches the nip point (the beginning of the consolidation zone). The top surface of the tape cools down faster than the bottom side due to direct contact with the roller. When the tape reaches the nip point, the top and bottom surfaces are at 383 °C and 375 °C, respectively. However, as soon as the tape enters the consolidation zone, the bottom surface of the tape quenches to the mold temperature rapidly (Fig. 14b). This causes the bottom surface to stay above the melting temperature for only 2.5 ms. On the other hand, the top surface stays above the melting temperature for 15 ms, a considerably longer duration.

The degree of intimate contact predictions based on the calculated temperature history of the bottom side of the tape are shown in Fig. 15 along with the experimental results. Similar trends were observed for the fractal and uniform rectangular intimate contact models. Despite the increase in pressure, almost no change occurs in the predictions. Due to the very limited time spent above the melting temperature at the bottom surface of the tape, the surface asperities do not flatten and the final degree of intimate contact calculation is dominated by the initial
surface parameters given in Table 2. The results are, therefore, only shown for the slowest placement speed (100 mm/s) since the predictions for higher placement speeds would result in the same final degree of intimate contact.

5. Discussion

Based on the results of surface and cross-sectional microscopy, it can be stated that intimate contact development during LAFP is a combination of multiple mechanisms. Squeeze flow of surface asperities, the common approach to explain intimate contact development for thermoplastic composites [19,20,22], plays a partial role. As seen in Fig. 10, samples manufactured with lower compaction forces contain incompletely deformed surface asperities filled with a mixture of fiber and resin. As the material is processed with higher compaction forces,
100 mm/s. L & S: Uniform rectangular surface representation [20], Y & P: calculated intimate contact based on calculated thermal history. Placement speed: direction [43,44]. Fig. 16 schematically depicts the proposed effective intimate contact development.

In Fig. 16a, the state of the thermoplastic composite tape prior to the nip point is shown. Zones 1 and 3 are fiber-rich regions, whereas Zone 2 represents the resin-rich portions of the material. After compaction (Fig. 16b), all of these regions are in physical contact. However, Zone 1 is not in effective intimate contact since no resin is present at the interface. Zone 2 achieves effective intimate contact with the squeeze flow mechanism. Zone 3 requires through thickness and in-plane percolation flow to establish effective intimate contact.

The share of these separate mechanisms on effective intimate contact development depends on the initial fiber-resin distribution of the tape and severity of microstructural changes during the heating phase of LAFP. Initial fiber-resin distribution of a thermoplastic composite tape highly depends on the tape manufacturing processes. Although similar constituents and specifications are provided for tapes produced by different manufacturers, fiber uniformity, resin content at the surface and void size/distribution varies [45–47]. In connection with the initial state of the material, the heating phase affects tapes manufactured by different suppliers differently, as reported in [28]. Also, the state of the substrate at the nip point should be considered for an accurate analysis of the final degree of effective intimate contact at each interface in a fiber placed laminate, which is beyond the scope of this work. Therefore, more efforts should be spent on understanding the microstructure of the thermoplastic tape and substrate just before the nip point during LAFP.

The response of the experimentally obtained degree of effective intimate contact to increasing compaction pressure differed from the response of the degree of intimate contact predicted by Eqs. (1) and (2), which use the squeeze flow assumptions. Intimate contact development based on homogenized transverse squeeze flow models were not sensitive to an increase in pressure. This insensitivity can be explained by the fact that only the surface temperature was used for predicting intimate contact development with the squeeze flow models. With the quenching effect of the tool surface, the resin near the surface is expected to cool down rapidly. However, molten resin is present through the thickness of the tape for a longer duration and contributes to the effective intimate contact development via percolation flow. Such a mechanism can also explain bonding below melting temperature, reported for CF/PPS tapes welded onto carbon woven fabric reinforced PPS laminates [48] and CF/PEEK laminates manufactured with LAFP [27]. This phenomenon was attributed to the autohesion of the amorphous chains in the semi-crystalline polymer at temperatures between the melting and glass transition temperatures. However, bonding was found to be limited to the intimate contact development in both works, indicating that its effect on the inter-laminar bond development was more pronounced than the autohesion mechanism. This means that the effective intimate contact development concept, which is introduced in this paper, might improve the accuracy of bonding predictions.

The samples manufactured with different placement speeds at a
fixed compaction force can be linked to different stages of intimate contact development. Fig. 17 shows effective intimate contact development as a function of compaction time, calculated by dividing the contact length obtained from pressure sensitive films by the placement speed. Experimental data for intimate contact development available in the literature covers a range from tens to hundreds of seconds [19,20,22]. Since only a fraction of a second is available for compaction during LAFP, it is difficult to make a direct comparison. However, the
trends show resemblance in terms of a steep increase at the beginning of compaction, followed by a decrease in the evolution rate. The same behavior was also confirmed for small time scales by numerical calculations presented in [49]. Two reasons can be identified as the reason for this behavior. The first one is the squeeze flow mechanism, which occurs at the resin-rich regions of the surface. Initially, the surface asperities have less contact and high local pressure, causing a rapid increase in the degree of effective intimate contact. As the asperities deform, increased contact area decreases the local pressure. Therefore, the evolution of the degree of effective intimate contact slows down. The second mechanism is the cooling of the tape as demonstrated in Fig. 14. The drop in temperature plays a hindering role for both the squeeze flow and percolation flow mechanisms as it causes the polymer viscosity to increase.

6. Conclusion

Intimate contact development during LAFP is a complex process involving multiple mechanisms. Due to short compaction times and very limited time available above the melting temperature of the semi-crystalline polymer, complete resin flow cannot be established at the tape surface as in conventional processing methods such as press molding and autoclave. Dry fibers were observed at the material surface after compaction, meaning that complete mechanical contact with the opposing surface might not contribute to the elimination of the interlaminar voids or development of bonding strength. In order to quantify the amount of flattened resin content at the surface, the concept of effective intimate contact was proposed and a methodology to obtain the degree of effective intimate contact from surface micrographs was presented.

Effective intimate contact develops with squeeze flow at the resin-rich portions of the surface; however, through-thickness and in-plane percolation flow is required to impregnate the regions with compressed dry fibers. Due to the percolation flow mechanism playing a role, the through-thickness temperature distribution of the tape gains importance for effective intimate contact development during the LAFP process. Currently available intimate contact models could not predict the experimentally obtained degree of effective intimate contact since they consider the material surface as a homogeneous fiber-resin mixture which deforms only with the squeeze flow mechanism. In addition, the through-thickness temperature distribution was not accounted for. Future work is identified as modeling effective intimate contact development with all contributing mechanisms and performing experiments for model validation.

CRediT authorship contribution statement

Ozan Celik: Conceptualization, Methodology, Software, Formal analysis, Investigation, Visualization, Writing - original draft, Writing - review & editing. Daniel Peeters: Conceptualization, Writing - review & editing, Supervision. Clemens Dransfeld: Conceptualization, Writing - review & editing, Supervision. Julie Teuwen: Conceptualization, Methodology, Writing - review & editing, Supervision.

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