EXPERIMENTAL INVESTIGATION OF INTER-LAYER THERMAL CONTACT RESISTANCE AND ITS RELEVANCE FOR CONSOLIDATION OF THERMOPLASTIC COMPOSITES

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Abstract

Carbon fiber reinforced plastics with thermoplastic matrices have become more important for industrial applications. A key factor for efficient processing of CFRP with thermoplastic matrix is the heating and cooling process of the material during consolidation. This process is governed by thermal conductance within the material on the one hand and by inter-layer thermal contact resistance on the other hand. In this paper we present an experimental setup as well as an evaluation method to calclulate the contact resistance between several layers. In comparison to unconsolidated samples, perfectly consolidated samples show a much better thermal conductivity. This indicates that contact resistance between unconsolidated layers does significantly decrease the overall thermal conductivity of samples which consist of multiple unconsolidated layers. Consequently, the time which is needed for heating processes increases. We can conclude, that interlayer-contact resistance is a phenomenon which must be considered, for the correct modelling of heating and cooling processes during manufacturing.

1. Introduction

Carbon fiber reinforced plastics (CFRP) with thermoplastic matrices have gained acceptance during the past years. Especially, the possibility of processing organosheets and tapes has opened up ways for new manufacturing processes such as press forming and automated tape placement. In all these processes several layers of prepreg with thermoplastic matrix are bonded to one laminate. In order to design efficient processes with thermoplastic materials, the bonding process ought to be fundamentally understood.

Three consecutive phases can be distinguished during the bonding process [1]: First, the layers are heated; second, the layers are consolidated and formed; third the laminate is cooled (Figure 1).

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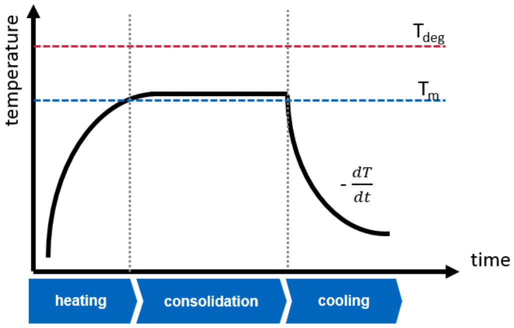

Figure 1. Different phases of bonding process.

Temperature is clearly the most influential parameter during the process. For the numerical optimization of the heating and cooling process and the accurate prediction of the temperature field thermal material properties such as thermal conductivity need to be determined. Besides the heat-flow within layers, which can be described by the thermal conductivity of the tapes, the thermal contact resistance between separate layers due to poor contact during heating must be considered. Figure 2 illustrates the thermal conductivity within layers and the contact resistance $R_{contact}$ due to poor contact between layers. As contact between separate layers improves during the consolidation process, thermal contact resistance of the complete laminate is also changing over time (Figure 2). In previous research [4] the inter-layer contact resistance of partially consolidated samples was determined by laser flash analysis (LFA) measurements. However, in our study we aim to measure the contact resistance of completely unconsolidated layers under a constant consolidation pressure which is not possible within a LFA.

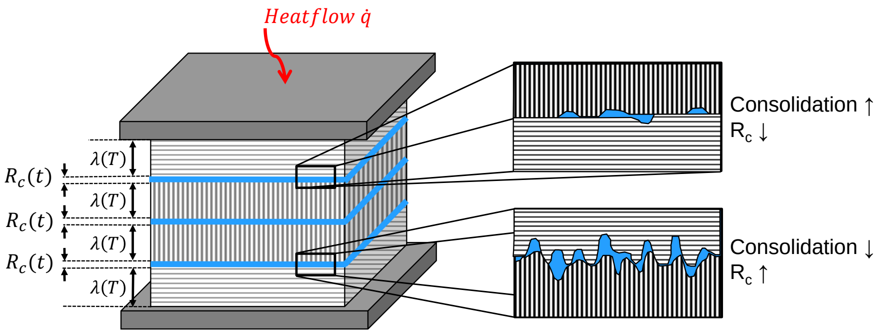

Figure 2. Principle of inter-layer contact resistance for thermoplastic tapes. Thermal contact resistance due to gaps at the contact surface between layers is marked blue.

Therefore, in this paper we will address the following questions: How can the thermal inter-layer contact resistance $R_{contact}$ between unconsolidated layers be determined experimentally? What is the relevance of inter-layer thermal contact resistance for heating of prepregs with thermoplastic matrix?

In order to answer these questions, we conducted heating experiments with thermoplastic-prepregs and logged the temperature-profile over time. We then simulated the experiments with a numerical simulation. By comparing the results from the simulation with experimental data, we quantified the thermal contact resistance for unconsolidated and consolidated samples.

In the first section of this paper we present some fundamental thermal properties of the material, which was investigated in this study. In the following sections we will present the approach, which was used to determine the inter-layer contact resistance in detail. This includes the experimental setup, the simulation , and the procedure to calculate the inter-layer contact resistance from these simulation results.

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We conclude this paper, by discussing the obtained results and the relevance of our findings for heating and cooling processes. The deeper understanding of the thermal contact resistance and its change during consolidation will allow for a more accurate prediction of heating and cooling times.

2. Materials

For this study carbon fiber reinforced prepregs with Polyamid-6 (PA-6) matrix (B3WC12) provided by BASF were used. The material has a nominal fiber weight content of 60%. The tape thickness is d=0.17 mm. The density ρ of the material is 1.46 g/cm^3.
For the evaluation of inter-layer contact resistance, which will be explained in the following section, the temperature-dependent thermal conductivity in transverse direction and the temperature-dependent heat capacity c_p of the material was determined. All samples were dried for 24 hours at 80 °C under vacuum prior to testing.
The thermal conductivity in off-plane direction (z-direction) of a single tape layer was measured by LFA. For the measurement samples of 20 mm x 20 mm were cut from the tape and coated with a very thin graphite film in order to prevent undesired reflection from the laser. The temperature of the samples was varied using a convective oven which was part of the LFA employed in this study. Due to the restrictions of the employed equipment the thermal conductivity was measured at discrete temperatures between 25 °C and 200 °C, which is the melting temperature of PA-6. As the result of the measurement the LFA provided the thermal diffusivity a. In general, the thermal conductivity λ is calculated, using the following expression:

$$\lambda = \rho c_p a \tag{1}$$

The heat capacity is determined by differential scanning calorimetry (DSC). Our measurements were conducted according to DIN 51007. The heat capacity was determined between 25 °C and 300 °C, which is approximately the degradation temperature of PA-6. The results of the LFA and DSC measurements are summarized in Figure 3.

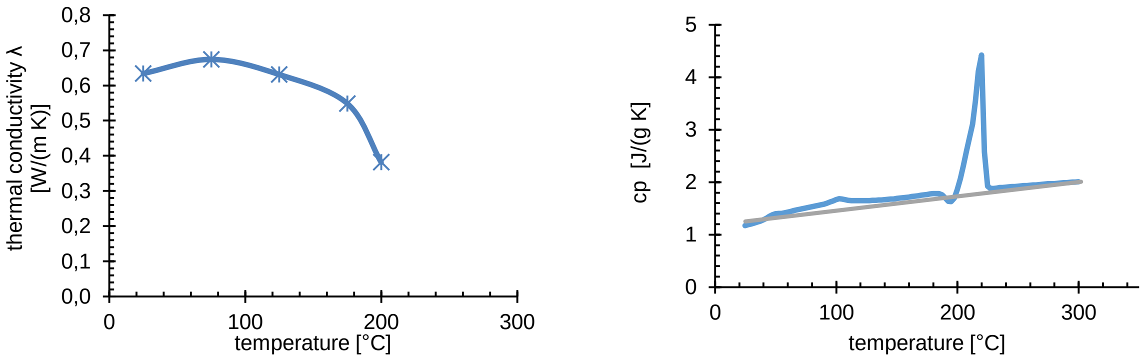

Figure 3. Thermal conductivity in off-plane direction of a single tape layer (left) and specific heat capacity of tape material (right)

The baseline of the heat capacity as a function of temperature, which is shown in Figure 3, can be approximated by the following linear expression:

$$c_p(T) = 0{,}0027\, T[°C] + 1{,}25 \tag{1}$$

3. Method

In this section of the paper we will explain the method, which was used to determine the inter-layer contact resistance. This procedure consists of three main steps: first, the experimental measurement of the temperature development in the transverse direction of thermoplastic tapes, second, the modelling

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and simulation of this heat-flow, and third, the evaluation of contact resistance, by comparing the experimental data with the results of the simulation. The evaluation of contact resistance is performed, using an iterative optimization approach. For the modelling of the heat flow, the material properties, which were described in the previous section, were used. The experiments were conducted using stacks of thermoplastic tapes. The entire procedure is illustrated in Figure 4 and the three main steps will be explained in more detail in the following sections.

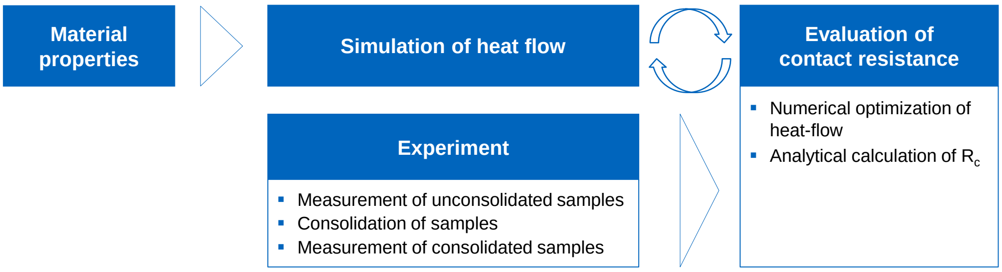

Figure 4. Overall scheme of method to determine the inter-layer contact resistance.

3.1. Experimental setup

The objective of the experiments was to measure the temperature distribution in thickness direction (z-direction) of the samples, which consist of several layers of tape material (see Figure 5). Samples were prepared by stacking 14 layers in a 0°/90° layup. The total sample thickness d_{Sample} was approximately 2.3 mm and the dimensions of the sample was 90 mm x 90 mm. Eight thermocouples (TC) of type K with a wire diameter of 0,08 mm were used to measure the temperature between layers in each sample. These micro thermocouples were employed to minimize the influence of the wiring on the contact between layers.

One of the employed thermocouple was placed underneath the first layer and one on the topmost layer. The remaining thermocouples were placed in between every other layer. The thermocouples were fixed to the surface of the tape material with a soldering iron.

The prepared samples were then placed on the lower plate of a temperature controlled press. A weight was placed on the sample to ensure a stable thermal boundary condition, constant pressure and a stable contact between the layers during the experiment. In the presented experiment a pressure of 400 kPa was applied by closing the press in addition to the gravitational force of the weight.

The experiment itself consisted of three phases: first each sample was heated three times in the unconsolidated state. During each measurement the lower plate of the press and the sample temperature were first equilibrated at 55 °C to allow for reproducible experimental conditions. Afterwards, the lower plate of the press underneath the sample was heated at a constant heating rate of 15 °K/min from 55 °C to the target temperature of 150 °C. The temperature development in the sample was logged with a sampling rate of 10 Hz. Then the sample was consolidated at a temperature of 250 °C for 10 minutes and cooled with a cooling rate of 15 °K/min. After consolidation we could assume a very good contact between layers. Then the identical consolidated sample is heated up again three times under the same conditions as the unconsolidated sample.

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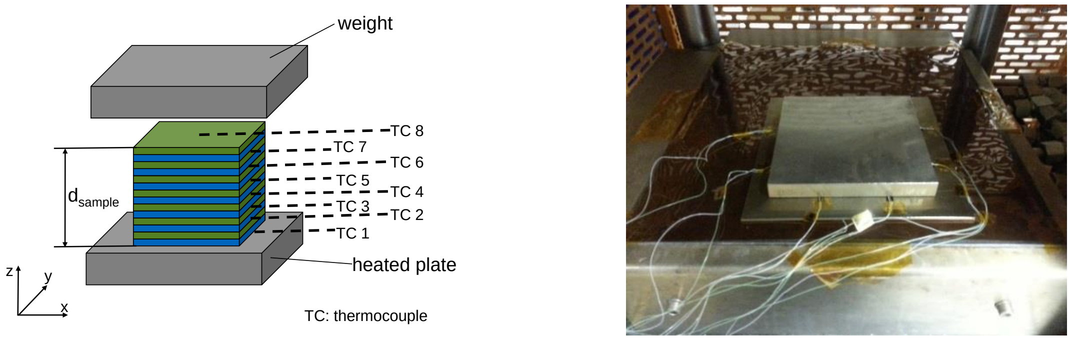

Figure 5. Experimental setup for a 2 mm thick sample, consisting of 14 individual layers and measurement of temperature profile with 8 thermocouples (left). Photo of setup (right).

3.2. Simulation of heat transfer

In order to differentiate the influence of inter-layer contact resistance from thermal conductance during the heating process of the samples a numerical model was set up.

The temperature distribution in the samples can be described by Fourier's law [3]:

$$\rho\, c_p \frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left(\lambda \frac{\partial T}{\partial x}\right) + \frac{\partial}{\partial y}\left(\lambda \frac{\partial T}{\partial y}\right) + \frac{\partial}{\partial z}\left(\lambda \frac{\partial T}{\partial z}\right) + \dot{\omega} \tag{2}$$

We assume, that each layer of the sample is heated homogenously in the x-y plane and the gradient in thickness direction (z-direction) is much larger than the temperature difference in the plane. Furthermore, there are no heat sources $\dot{\omega}$ in the laminate. Then the general form of Fourier's law can be simplified to [3]:

$$\frac{\rho\, c_p}{\lambda} \frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial z^2} \tag{3}$$

As there is no general solution for equation (3), it can only be solved numerically. Therefore, an implicit finite volume model was implemented in Matlab®. Each tape layer was modelled by 14 volume elements and one time step is 0.1 s. As boundary conditions the measured temperature of the press plate (TC 1) was used for the first elements. For the topmost element a heat sink boundary condition was implemented, which accounts for the heat capacity of the steel weight on top of the sample. The magnitude of the heat sink was determined from the experiments with the consolidated samples.

In order to determine the contact resistance of the unconsolidated sample, we first aimed at determening the equivalent thermal conductivity λ_{equ} of each sample. The equivalent thermal conductivity is a smeared value, which accounts for the thermal conductivity of each layer and for the additional inter-layer contact resistance between layers (see Figure 6). This value represents the overall thermal conductivity of each sample, which consisted of k layers and i=k-1 contact surfaces between layers. A priori λ_{equ} was not known. The identification of λ_{equ} and the calculation of the contact resistance will be described in the next section.

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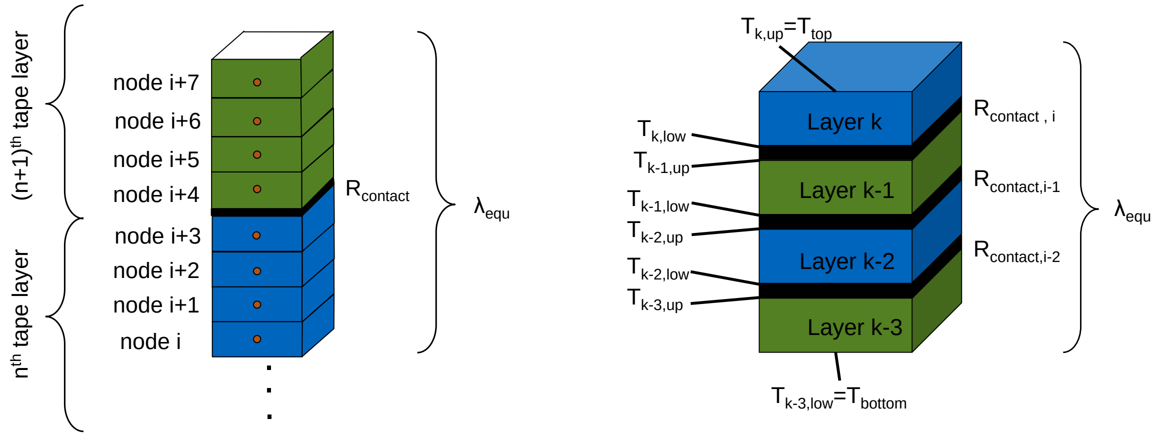

Figure 6. Illustration of 1D numerical model (left). Thermal model for determination of R_C (right)

3.3. Evaluation of inter-layer contact resistance

In the previous sections, we discussed inter-layer thermal contact resistance as a phenomenon, which occurs due to incomplete contact between single layers. In this section we will describe the method, which was used to quantify inter-layer contact-resistance. This method consists of two steps. First, the equivalent thermal conductivity λ_{equ} of each sample had to be calculated, and second, the inter-layer contact resistance R_C was evaluated from λ_{equ}.

In order to determine λ_{equ} for the investigated samples an optimization scheme was implemented in Matlab®. This scheme compares the experimentally determined temperature distribution of m thermocouples with the respective results from the simulation. The optimization scheme uses a least square method, which minimizes the error between the experimental temperature and the simulated temperature:

$$min\left(err(\lambda_{equ})\right) = min\left(\sum_{m}[T_{Exp}^{m} - T_{Sim}^{m}(\lambda_{equ})]^2\right) \tag{4}$$

For the evaluation of R_C we followed the idea of [2] and generalize their approach. In Figure 6 the sample is modelled as a series of k tape layers, which are separated by i=k-1 interfaces. According to [2,4], the heat flux in each tape layer as well as in each resistive layer is equal. The heat flux $\dot{q}$ across the kth tape layer of the sample (see Figure 6) can be calculated from the temperature difference between the upper and the lower surface of the layer, the thermal conductivity of one tape layer $\lambda_k(T)$ and the thickness of one layer d_k by the following equation:

$$\dot{q} = \frac{\lambda_k(T)}{d_k}(T_{k,up} - T_{k,low}) \tag{5}$$

Similarly the heat flux across the resistive layer is calculated according to equation (6).

$$\dot{q} = \frac{(T_{k,low} - T_{k-1,up})}{R_C} \tag{6}$$

Assuming, that each sample is composed of k tape layers with identical thickness d_k we can write the heat flux across the sample as:

$$\dot{q} = \frac{\lambda_{equ}}{k\ d}(T_{top} - T_{bottom}) \tag{7}$$

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As the change in thermal conductivity in the temperature range between 55 °C and 140 °C is small the thermal conductivity $\lambda_k(T)$ of each layer can be assumed to be constant. Combining equations (5), (6) and (7) yields the final expression for the inter-layer contact resistance:

$$R_C = \frac{k\,d}{k-1}\left(\frac{1}{\lambda_{equ}} - \frac{1}{\lambda}\right) \tag{8}$$

4. Results and Discussion

In this paper we pursued two goals: the development of an experimental approach to determine the inter-layer contact resistance of thermoplastic tape materials before and after consolidation and we wanted to discuss the relevance of this phenomena for processing of PA-6 tapes, based on the findings of our experiments.

Before evaluating the quantified value for the inter-layer contact resistance, we analyzed the experimental raw-data, namely the temperature development over time in the samples. Figure 7 shows the temperature distribution in the sample for different points in time. The diagram on the left shows the results for an unconsolidated sample and the diagram on the right side the temperature distribution for a specimen after consolidation. At the beginning of the experiment no temperature gradient can be observed. After 240 s the slope of the temperature curve increases, as the temperature of the press plate is increased for the unconsolidated sample. A constant slope is reached after approximately 480 s. When comparing the consolidated sample to the unconsolidated specimen, it can be observed that the slope of the temperature curve is much flatter. This implies, that the temperature gradient between the first layer of the consolidated sample and the top most layer is very small. The small temperature gradient indicates a very good thermal conductivity across the sample thickness. One can conclude that the overall thermal conductivity across the sample thickness direction, which we previously described as λ_{equ} has been improved during consolidation. As the thermal conductivity $\lambda(T)$ of the single tape layers remains constant, the thermal contact resistance between layers must have been decreased during consolidation.

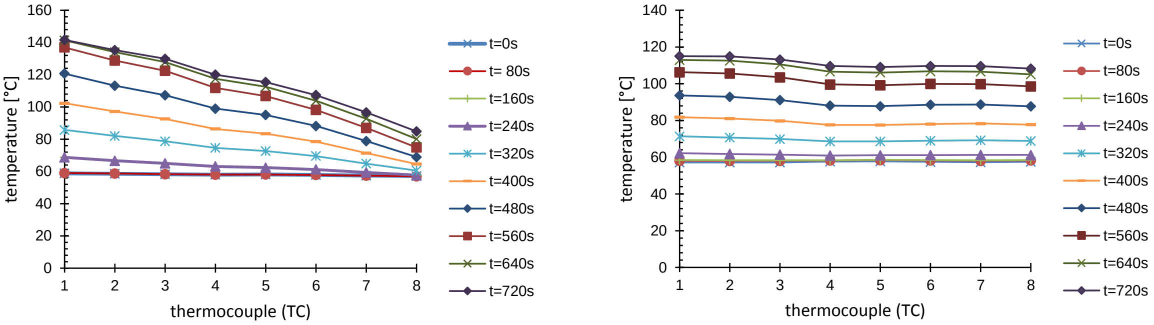

Figure 7. Temperature development in the sample over z for an unconsolidated sample (left) and a consolidated sample (right)

During the evaluation of λ_{equ} of the consolidated samples, using the described optimization process, 30% of the samples had to be excluded because of faulty temperature measurements. We assume that during the consolidation process thermocouples were released or pressed into the sample. In these cases the optimization scheme ended at the upper or lower boundary value. This was one of the major draw-backs of the developed setup. Also, the small temperature gradient in thickness direction, which was observed for the consolidated samples in some cases, led to bad fitting results. To ensure the quality of the fit the rout mean square error between the optimized temperature distribution and the measured temperature field was calculated. Results with R^2<0.9 were excluded from the evaluation. Whereas the 30% of the consolidated samples had to be excluded according to this criterion all measurements with unconsolidated samples could be evaluated.

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For the calculation of the contact resistance according to equation (8) we used the mean value of 0.65 W/(mK) for the thermal conductivity of the tapes in the temperature range between 25 °C and 150 °C. The results of the evaluation are summarized in Table 1. The inter-layer thermal contact resistance between layers of unconsolidated sample is 36 times higher than that of perfectly consolidated layers. Comparing our results with the findings of Levy et al. [2], who linked contact resistance to the degree of intimate contact, we can see that the results match quite well. We recall that the degree of intimate contact is defined as the surface in contact of two adjacent layers divided by the total surface area of theses layers [5]. For well consolidated samples with a degree of intimate contact of 0,825-0,940 they determined values of $0.75\text{x}10^{-4}$ and $0.9\text{x}10^{-4}$ $(\text{m}^2\text{K})/\text{W}$ for inter-layer contact resistance. For the perfectly consolidated samples in our experiment with a degree of intimate contact of approximately more than 0.95, we obtained only slightly higher results. The completely unconsolidated samples of our experiment have a 10 times larger contact resistance than the partially consolidated samples (degree of intimate contact 0.65) presented in [2].

Table 1. Thermal contact resistance evaluated for thermocouples 1-8 (k=13).

Specimen	equivalent thermal conductivity λ_{equ} (W(mK)^{-1})		thermal contact resistance R_C $(\text{m}^2\text{K})\text{W}^{-1}$	
	mean	standard deviation	mean	standard deviation
consolidated sample	0.482	0,082	$0.988\text{x}10^{-4}$	-
unconsolidated sample	0.047	0.004	$36.45\text{x}10^{-4}$	-

The results show, that the presented experimental approach is suitable to determine the inter-layer contact resistance between different layers of thermoplastic tape material. Compared to LFA measurements, completely unconsolidated samples can be investigated with a varying compaction pressure. The obtained values for contact resistance show, that the overall thermal conductivity is significantly decreased due to poor contact between different layers. For the majority of the heating period, up to the melting temperature of common thermoplastic materials the thermal contact resistance is governing the heating process. Therefore, the phenomenon of thermal contact resistance significantly influences the consolidation process.

5. Conclusion and Outlook

In this paper we presented an experimental procedure, which is suitable to quantify the inter-layer thermal contact resistance of thermoplastic tapes before and after consolidation. The experimental method allows the characterization of contact resistance under realistic processing conditions.

The equivalent overall thermal conductivity of an unconsolidated 14 layer sample decreases by a factor of 10 compared to a perfectly consolidated sample. Therefore, we can conclude that for the precise prediction of heating times during consolidation in a temperature controlled press, contact resistance must be accounted for. Furthermore, it must be noted that contact resistance is reduced during consolidation. Therefore, the thermal material properties for heating processes before consolidation differ from thermal material properties during cooling of the material after consolidation.

Future research should explore the influence of compaction pressure on thermal contact resistance as well as the correlation between the tape's roughness and the inter-layer thermal contact resistance.

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