THE INFLUENCE OF MOISTURE ON THE DE-CONSOLIDATION BEHAVIOUR OF CARBON FIBER REINFORCED PA-6 LAMINATES

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Abstract

Carbon fiber reinforced thermoplastics offer the possibility to use fully consolidated semifinished products for thermoforming or fusion bonding processes. Nevertheless, processing above the melting temperature of the polymer may increase the porosity due to de-consolidation. The degree of de-consolidation depends on different factors such as fiber volume fraction, layup, pressure and moisture within the composite. The influence of moisture on the de-consolidation behaviour of carbon fiber reinforced Polyamide-6 is investigated using pressure dependent de-consolidation experiments. De-consolidated samples are characterized by X-ray computed tomography and optical microscopy. Analysis of the measurements show a strong influence of the moisture content on the de-consolidation behaviour. Effects of de-consolidation can be reduced using dry samples or pressures in the kPa range.

1. Introduction

De-consolidation describes the loss of consolidation during heat treatment of the fiber reinforced thermoplastics above the melting temperature [1]. This effect increases the porosity of the composite during processing steps such as thermoforming [2] and fusion bonding [2–4] and reduces the mechanical performance of the component. Experimental studies show the development of elliptical voids up to a few hundred micrometers [4, 5, 1] located in matrix rich areas and additional longitudinal voids within fiber rovings [1]. The increase in porosity is typically in the range of 10% [1] -12% [5], but also values up to 72% are reported [6].

The degree of de-consolidation depend on different microstructural properties and processing parameters. Henninger [1] and Shi [4] showed a decrease of de-consolidation with increasing pressure. Additionally, both authors found a critical pressure > 1 MPa from which the development of additional voids is suppressed. An increase in temperature accelerates the development of voids but does not increase the amount of de-consolidation [1, 7]. Wan [8] and Gröschel [5] reported a higher degree of deconsolidation with increasing fiber volume content. This is also the case for a higher moisture content within the thermoplastic matrix [5] and an increase in pressure during consolidation [9].

According to Ye [10] the driving force of de-consolidation is a combination of three different effects: coalescence of existing voids, increase of gas volume due to thermal expansion and release of elastic energy which is stored within the fiber network. Since coalescence of existing voids does not increase the total amount of porosity and an increase of void volume due to thermal expansion is not capable to explain a total void volume in the range of 10%, so the release of stresses within the fiber textile is considerd as main driving force for de-consolidation [10].

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The amount of elastic energy stored in the fiber network depends on the layup of the composite. Experimental studies for short fiber reinforced composites and woven fabrics are known for different material combinations such as glass and carbon fibers in combination with Polyether-ether-keton (PEEK), Polyether-sulfone (PES) or Polyamide (PA) matrix. Nevertheless, there is a lack of experimental results concerning non-crimp carbon fiber reinforced Polyamide-6 (PA-6) laminates, especially with regard to the initial moisture content during processing.

In this study we investigate the pressure dependent increase of the porosity in carbon fiber reinforced PA-6. Therefore, we induce de-consolidation via heating the composite above the melting temperature of the polymer. During the heat treatment different pressures from 10 Pa to 4000 Pa were applied in a compression setup. To reveal the morphology and the void content microscopic and X-ray computed tomography (CT) analysis is performed.

2. Experimental

2.1. Material and sample preparation

A carbon fiber reinforced PA-6 $[(0/90)_3]_{sym}$ laminate with a thickness of 2 mm was provided by SGL Carbon GmbH (Meitingen). The laminate was manufactured from pre-impregnated tapes using a heating press. Microscopy pictures show a void free laminate at magnifications up to 500x. According to void content estimations from CT measurements the volumetric void content is < 0.5%.

Samples of 20 x 20 mm were cut from the laminate using a water-cooled diamond saw. Samples denoted as “wet” were stored in a climatic chamber (Klimaprüfschrank WK3-340/70, Weiss Umwelttechnik GmbH) at 70°C and 62%RH according to DIN EN ISO 1110 until mass equilibrium was reached. “Dry” samples where dried in a vacuum oven at 80°C until the mass decrease was smaller than 0.05% within 3 days.

De-consolidation experiments were performed via a DMA (Q800, TA Instruments) using compression plates. A constant pressure between 10 Pa and 4000 Pa was applied during the de-consolidation experiments. The temperature profile starts by heating the specimen to 240°C with a heating rate of 10 K/min followed by a 15 min isothermal heating. Subsequently, the specimen was cooled without active cooling to room temperature with a maximum cooling rate of 10 K/min at 240°C. The thickness of the specimens was determined before and after the heat treatment using a micrometer gauge. To avoid adhesion between the compression plates and the specimens a release agent was used.

2.2. X-ray computed tomography

CT was used to get an insight into the material and for estimating the void content within the samples. X-ray images were collected using a Nanotom m180 (GE Sensing & Inspection) applying an electron current of 135 µA and an acceleration voltage of 100 kV. The images were taken using a magnification factor of 8.6 (voxel size ~11.7 µm). For reconstruction the datos|x2 reconstruction software package was used.

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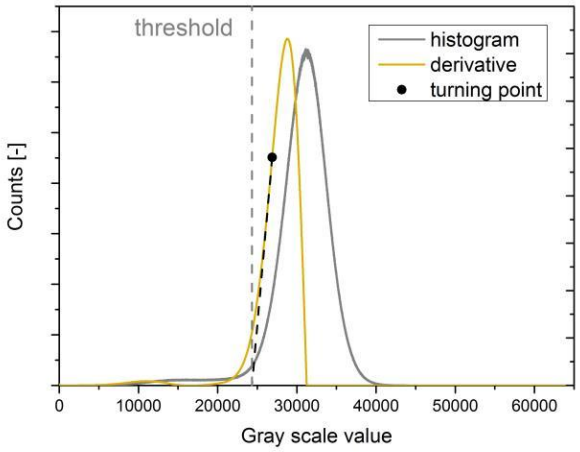

Figure 1. Procedure to determine threshold value between voids (left) and material (right)

Furthermore, estimation of the void content from the reconstructed volumes was performed using a threshold procedure. Therefore, the grayscale histogram within the sample boundaries was determined. The threshold was calculated by the intersect of the grayscale value axis and the tangent of the second derivative at the turning point of the material's peak as shown in Fig. 1. The analysis was executed independently for each sample.

2.2. Optical microscopy

Microscopy images were prepared to resolve the resolution limitations of CT. Therefore, the samples were embedded in epoxy resin and subsequently grinded and polished. Images were taken using a Keyence VHX-2000 digital microscope. Optical microscopy is highly suited to detect pores and voids in the micrometer range, especially in fiber rich regions.

3. Results and Discussion

Both, dry and wet samples showed an increase in thickness (>0 - 10%). Visually the quality of the surface decreases due to an increase in surface roughness after the heat treatment. Only very small polymer droplets can be observed at the edges of the samples. Therefore, we can assume that only little squeeze flow of the polymer occurred. Depending on the conditional state of the samples and the pressure conditions voids can be spotted at the edges of the samples by eye.

Representative microscopy images of the polished samples are shown in Fig. 2 (a) - (c) for the dry de-consolidated samples at 4000 Pa, 200 Pa and 10 Pa, respectively. The microstructural features such as fiber volume content, thickness of individual layers and low void content are mostly unaffected by the heat treatment. Nevertheless, laminates at all pressure levels show a higher surface roughness compared to the initial state. Images at higher magnification factors show no additional voids within the fiber rich regions.

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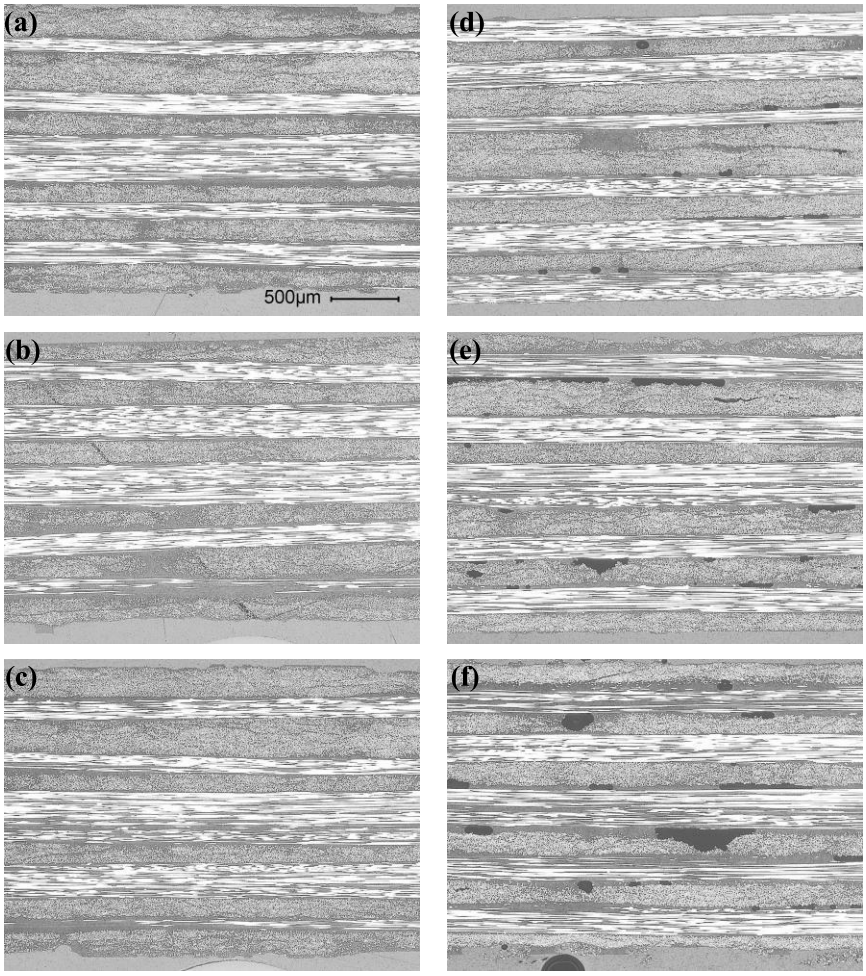

Figure 2. Cross-sections of de-consolidated samples in dry condition at a applied pressure of (a) 4000 Pa, (b) 200 Pa, (c) 10 Pa and in wet condition at applied pressures of 4000 Pa (d), 200 Pa (e) and 10 Pa (f).

Cross-sections of samples de-consolidated in wet condition are shown in Fig. 2 (d) - (f) for 4000 Pa, 200 Pa and 10 Pa. At 4000 Pa the samples show voids up to 150-200 µm. The voids are located within fiber rich regions at the boundary between two adjacent layers and have both. spherical and elliptical shapes. With decreasing pressure during the heat treatment the total amount of voids increases. The voids grow substantially along the boundary of the layers and form delamination like defects at length scales in the milimeter range. Additionally, smaller voids can be found within the fiber rich regions in the layers themselves. At even lower pressures the voids grow in thickness direction and reach extensions in the length scales of the layers' thickness. Separation of layers located parallel to the fiber direction can be found at several locations. As described for dry samples, the surface quality is reduced due to an increase in surface roughness.

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The distribution of voids within the volume of a sample de-consolidated at 10 Pa in dry condition is shown in Fig. 3. In contrast to the cross-section images from microscopy, CT images reveal voids initiating at the edges of the sample. The voids are aligned parallel to the fibers and extend through a complete layer thickness. Nevertheless, the majority of the volume is void free. In comparison to this the sample de-consolidated at 10 Pa in wet condition contains many additional voids as shown in Fig. 4. Beside the large gaps within the layers, spherical voids up to several hundred micrometer can be observed and longitudinal gaps in the fiber direction are distributed over the whole volume. In addition, a macroscopic delamination of 4 x 4 mm can be found on the bottom right corner of the sample.

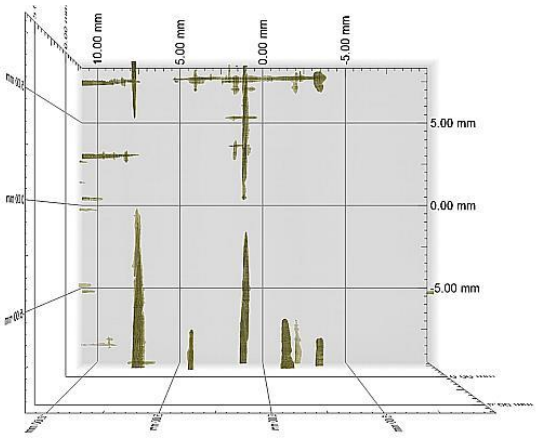

Figure 3. Void distribution within a sample de-consolidated at 10 Pa in dry condition.

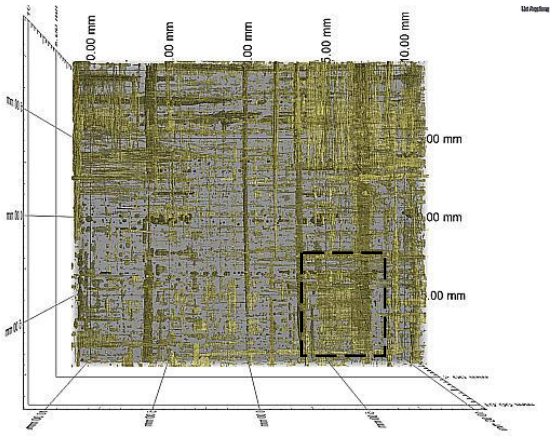

Figure 4. Void distribution within a sample de-consolidated at 10 Pa in wet condition (**- - -** delamination).

The porosity estimated from CT measurements as described in the experimental section is shown for dry and wet samples in Fig. 5. Dry samples show a constant void content of about 1% for all pressure levels from 10 Pa to 4000 Pa. As already shown in the microscopic analysis the void content of wet samples is higher after de-consolidation and increases from ~1% at 4000 Pa to 5% at 10 Pa. The void content is nearly constant from 10 Pa to 200 Pa whereas there is a huge drop between 200 Pa and 4000 Pa.

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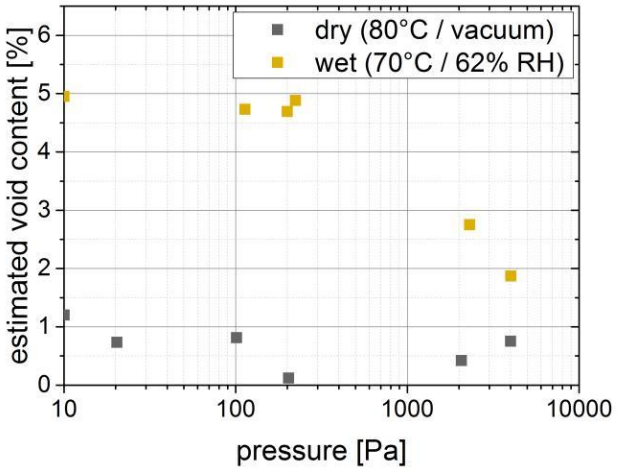

Figure 5. Void content from X-ray computed tomography of samples de-consolidated in dry and wet condition under pressure.

The results indicate that the elastic energy stored in the fiber network is not the main reason for de-consolidation of non-crimped carbon fiber reinforced PA-6 as suggested by Ye [10] for other material combinations and layups – especially wovens. The results suggest evaporation of water within the matrix in liquid state is a major cause of de-consolidation. Nevertheless, more complex fiber configurations may increase the amount of elastic energy stored within the fiber network and lead to a combination of both driving factors, as indicated by the results of Gröschel [5].

As described by other studies a critical pressure exists were de-consolidation can be suppressed. The critical pressure is in the kPa range which is similar to earlier studies [1, 4]. Therefore, especially processes without the ability of applying additional pressure such as back injection or processes with inhomogeneous pressure distribution may suffer from de-consolidation.

3. Conclusion

The results indicate a strong influence of moisture on the deconsolidation behavior of carbon fiber reinforced PA-6. Within de-consolidated wet (70°C, 62%RH) samples the void content is up to 500% of the void content within de-consolidated dry (80°C, vacuum) samples. Optical microscopic analysis and X-ray computed tomography indicate that voids agglomerate between laminate layers which leads to delaminations on large length scales.

The results suggest that drying of fiber reinforced PA-6 is crucial for production of void free components if homogenous pressure distribution above several kPa is not applicable. Process models describing the consolidation, thermoforming and fusion bonding should not neglect the influence of de-consolidation and moisture within the polymer matrix to provide reliable results.

Acknowledgments

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