Influence of Fabric Parameters on Surface Waviness of FRPC: An Experimental Investigation and Development of a Model on Surface Waviness

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Abstract
This paper deals with the surface development of continuous fiber-reinforced thermoplastic composites, so called organic sheets, during processing. To investigate the effect of the textile parameters on the surface development organic sheets with different steel wire fabrics and a polycarbonate matrix were manufactured using a laboratory press. The fabrics differed in fiber diameter and mesh size. The results showed that both textile parameters have a significant influence on the characteristic surface waviness of the FRPC. Increasing fiber diameters and mesh sizes lead to higher maximum waviness. Organic sheets with a fiber diameter of the reinforcement less or equal 50 µm could not be differentiated anymore and had no visible waviness. Based on the equation for linear thermal expansion a thermo rheological process model was developed to predict the surface waviness. The model describes the waviness creation including a pressure induced compensating polymer flow. A waviness comparison of experimental data with calculated results shows a good agreement.

Keywords: *Organic sheet, Surface waviness, Variothermal process, Polycarbonate, Polymer rheology*

1 Introduction

Lightweight fiber-reinforced polymer composites (FRPC) enable sustainable, innovative and intelligent solutions for a broad range of industries. E.g. short-fiber-reinforced polymers offer technically and economically advantageous solutions in various applications in high-volume industries such as the automotive industry. Approximately nine % of total vehicle weight, which is about 50% of the vehicles volume, are accounted for polymeric materials, most of which is however not fiber-reinforced [1]. Due to high material and process costs a high volume use of continuous fiber-reinforced parts in (semi-) structural applications is not yet established, despite its high lightweight potential [2]. A study of the Association of German Engineers (VDI) from the year 2014 explicitly highlights the potential through the use of continuous fiber reinforced thermoplastic semi-finished products, so-called organic sheets, for a more successful market penetration. Through the production of flat semi-finished products and the fast, subsequent thermoforming process in combination with an automated process chain, organic sheets provide a substantial cost reduction potential compared to thermoset methods [3-5]. Approximately 40% of the vehicle weight can be allocated to the car body structure. Through the consistent use of fiber-reinforced polymer composites, the body weight can be reduced by approximately 25% [6].

One of the biggest technological challenges for the use of organic sheets in high quality visible applications consists in the extensive effort in surface treatment, which is necessary for the achievement of the high optical requirements for class-A components. The varnishing produces up to 40% of the total component costs and thus becomes an important economic decision

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criterion [7,8]. Therefore, substrate surface qualities have to be optimized in order to achieve more competitive components. This can be done either by altering the laminate structure itself and/or by adapting the manufacturing process.

The majority of scientific work focuses on thermoset materials. A first theoretical model to calculate the fiber print-through of unidirectional reinforced thermosets was developed by Kia. It uses a mechanical analogous model to describe the heterogeneous shrinkage behavior of fibers and matrix. Kia also postulated that increasing the surface layer thickness is more effective than increasing the surface layer stiffness [9,10]. The influence of the matrix is topic to various publications. For thermosets it is known that a small chemical shrinkage as well as a high water uptake increase surface homogeneity [11-13]. When thermoplastic materials are used amorphous polymers such as polycarbonate lead to lower surface waviness than semi-crystalline polymers such as polyamide, due to the absence of crystalline shrinkage in case of amorphous polymers [14]. Information on the influence of fiber architecture is fragmentary. Blinzler found out that the type of fiber (glass or carbon) has only little influence on the surface properties [14]. Work on the influence of single filament diameters between 5 – 20 µm on the surface properties is diametric [15,16]. Schubel et al. investigated the effect of varying yarn counts. When using 2/2 twill fabrics with yarn counts from 3k to 12k an increasing roving diameter increases the surface roughness [12].

A large part of published literature uses roughness for the surfaces characterization of fiber-reinforced polymer composites (FRPC) [17-19]. According to standard DIN 4760 the periodicity of the surface structure induced by the textile reinforcement is defined as waviness, not roughness [20]. The waviness parameters are better suited to characterize the surface properties.

2 Materials

The choice of matrix material for the experiments presented in this work is based on a range of requirements. In addition to the suitability for high surface qualities, sufficiently high mechanical and thermal properties as well as an economically reasonable price are main criteria. Due to the low thermal shrinkage and the high continuous operating temperature amorphous polycarbonate Makrolon® 2408 is used (see Table 1). The melt flow rate of the polymer at 300°C and 1, 2 kg force is 20 g/10 min [21]. The polycarbonate (PC) does not contain any volatile additives such as, for example, release agents.

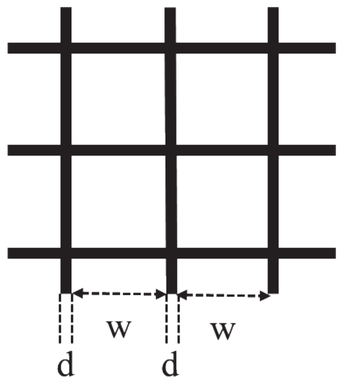
The PC has been modified with carbon nanotubes (CNT) Baytubes® C70P (one weight-%) to blacken the material in order to enable a correct optical characterization of the otherwise transparent PC. The CNTs are dispersed into the polycarbonate using twin-screw extrusion process and then calendared into films with a mean thickness of 100 µm.

Table 1: Material properties of Makrolon® 2408 including CNT

Density ρ	Young's-Modulus E	Coefficient of thermal expansion αth	Glass transition temperature Tg
[kg/m3]	[MPa]	[1/K]	[°C]
1206	2350	< 150 °C: ≈ 65 · 10-6 > 150 °C: ≈ 216 · 10-6	147

To systematically evaluate the influence of the textile parameters (roving diameter d and mesh size w) on the surface properties of organic sheets, conventional filament fabrics are of limited suitability. Due to the roving structure made of thousands of filaments embedded in a thermoplastic matrix and the plastic deforming ability during the processing distortion of the roving's cross section cannot be eliminated. The use of industrial steel wire fabrics, which are produced in close tolerance according to DIN ISO 9044, avoid variations in fiber diameter and mesh geometry, while covering a wide range of parameters.

The rovings consist of a single metal fiber and the mechanical characteristics do not change during processing. Therefore, the experimental study on the influence of fabric parameters is performed with 14 different steel wire fabrics with varying fiber diameter d and mesh width w with the parameters listed in table 2 (see Figure 1).



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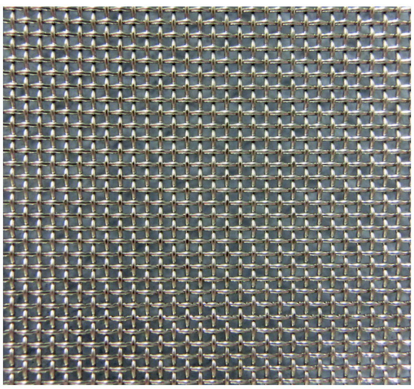


(b)

Figure 1: (a) Fabric parameters fiber diameter d and mesh size w, (b) Example of a plain weave steel wire fabric.

Table 2: Overview of the investigated fiber diameter and mesh size combinations

Mesh size w	Fiber diameter d	Theoretical number of matrix films to achieve a surface layer thickness of 100 µm
[µm]	[µm]	
25	25	2,32
50	40	2,61
63	40	2,55
77	50	2,48
100	70	2,96
200	125	3,57
250	200	4,54
315	110	3,84
315	160	3,96
315	200	4,9
315	250	4,89
400	250	5,0
500	320	6,19
1000	630	10,54

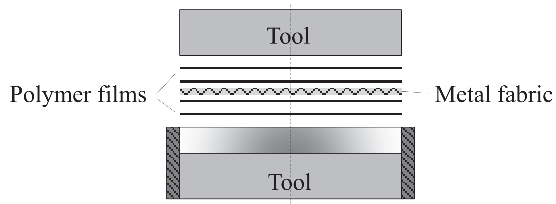


Figure 2: Schematic drawing of the cylindrical tool and a single reinforcement layer laminate build up.

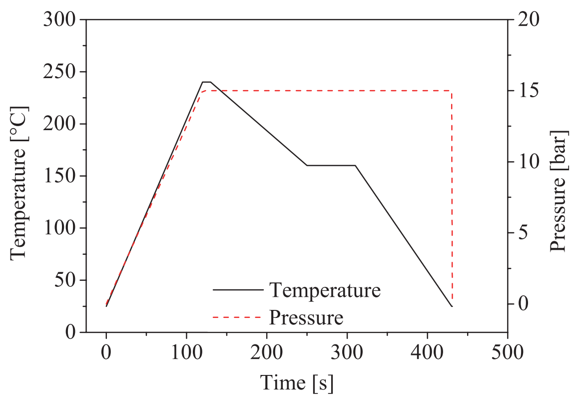


Figure 3: Process cycle to manufacture the organic sheets.

3 Methods

3.1 *Variothermal manufacturing*

Research on the influence of fabric parameters on the surface properties of organic sheets is carried out on a variothermal laboratory press. The associated press tool is a cylindrical steel tool with an inside diameter of 50 mm (see Figure 2).

The process cycle is designed with a focus to the cooling stage. As the fabrics are made of mono fiber steel wires only a macro impregnation occurs during the manufacturing of the organic sheets. This leads to a moderate processing temperature of 240°C and a total process time of 430 seconds. The isothermal holding phase at 160°C is used to reduce internal tension through relaxation, while holding the temperature above PCs glass transition temperature (see Figure 3).

The organic sheets are designed with only one layer of reinforcement to avoid multi-layer influences on the surface properties such as nesting. To investigate the surface waviness it is important to keep the surface layer thickness as constant as possible. The different fiber diameters result in different fiber volume contents for each incompressible fabric. Therefore, fiber volume content is not kept constant for the laminate configurations. The desired surface layer thickness is set to 100 µm. As the matrix is supplied as film material with a mean thickness of 100 µm the theoretical number of films needed to achieve the target 100 µm is listed in table 2. The values are calculated as the sum of polymer volume needed to completely impregnate the fabric structure and build a surface layer thickness of 100 µm. Based on the calculated values the number of films used is round up to a whole number. The redundant amount is for matrix squeeze out during manufacturing.

3.2 *Surface development in FRPC*

Existing models describing the surface structure of thermoplastic composite are based on the fact that locally varying thermal shrinkage between matrix-rich zones and fiber-rich zones, resulting from the different coefficient of thermal expansion (CTE) of fiber and matrix, causes a surface waviness. The amplitude of the waviness is dependent on the laminate structure in thickness direction. A cross section A-A is used for calculating the maximum thermally induced waviness (see Figure 4).

Due to the symmetrical design of a plain weave laminate, the calculation can be simplified by just considering the upper half of the cross-section. In addition, the elliptic fiber cross-section is simplified to a circular cross-section (see Figure 5).

The thermally-induced maximum waviness Wz can be calculated as the difference between the thermal expansion above the matrix-rich region and surface layer Δh_M and the fiber-rich region and surface layer Δd_F.

$$\Delta d_F = d_0\,\alpha_F\,\Delta T + h_1\,\alpha_M\,\Delta T \quad (1)$$

$$\Delta h_M = h_0\,\alpha_M\,\Delta T + h_1\,\alpha_M\,\Delta T \quad (2)$$

$$Wz = \left|h_0\,\alpha_M\,\Delta T + h_1\,\alpha_M\,\Delta T\right| - \left|d_0\,\alpha_F\,\Delta T + h_1\,\alpha_M\,\Delta T\right|$$

where $h_0 = d_0$ (3)

$$\Leftrightarrow Wz = \left|h_0\,\alpha_M\,\Delta T\right| - \left|d_0\,\alpha_F\,\Delta T\right|; \quad (4)$$

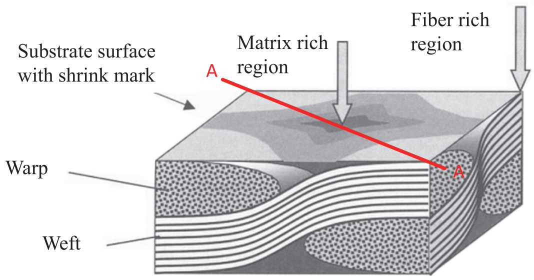


Figure 4: Cause of surface texture through local variations in fiber volume fraction and different coefficients of thermal expansion for matrix and fiber.

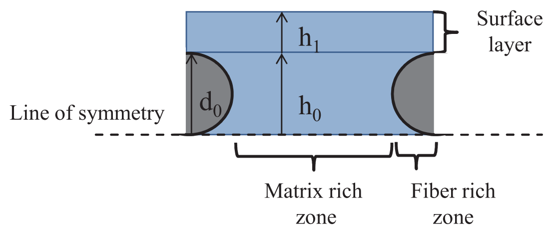


Figure 5: Idealized, diagonal cross-section A-A from Figure 4 with additional surface layer h_1.

with α_M = CTE of matrix
αF = CTE of fiber
h_0 = initial height of matrix-rich region
h1 = initial height of surface layer
d_0 = initial height of fiber-rich region
ΔT = temperature difference

Equation 4 show that the presence of a homogeneous surface layer does not affect resulting waviness for purely thermal shrinkage, as it exists in both fiber- and matrix-rich region. Nevertheless this simple thermal model allows an analytical assessment of the expected waviness in the composite. The most important simplifications of this one-dimensional model are:

- Assumption of a homogeneous surface layer
- Volumetric shrinkage is not taken into account
- No consideration of mechanical properties of components

3.3 *Surface characterization*

The characterization of surface properties is carried out through white light profilometry on clean, grease-free samples.

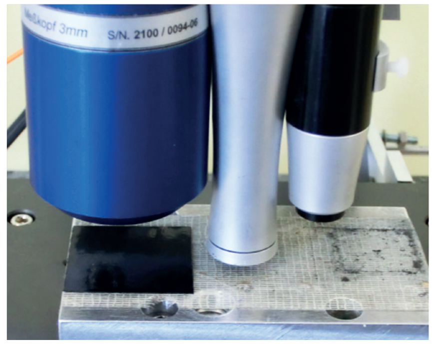
The specified values are mean values and standard deviations of the individual measurements. Due to material inhomogeneity and the small number of performed tests, the Nalimov outlier test with a confidence level of 95% is applied on the data, to make a secure statistical statement. The respective measured value x* is used in formula (5) and r* is calculated. The decision if an outlier is found is made by comparing this value with the corresponding value r_i from the r-table [22].

$$r^* = \frac{|x^* - \bar{x}|}{s} \cdot \sqrt{\frac{n}{n-1}} \tag{5}$$

with $\bar{x}$ = mean value
s = standard deviation
n = number of measurements
$r^* < r(95)$ = no outlier identifiable
$r(95) < r^*$ = presumably an outlier identifiable

If a value is identified as an outlier, it will be removed and mean and standard deviation are recalculated without the outlier value. After that the Nalimov test is repeated.

The surface topography is measured with a FRT MicroProf® white light profilometer. The used sensor has a lateral resolution of 5 - 6 µm and a vertical resolution of approx. 0.1 µm according to manufacturer's instructions. The waviness value is calculated on the basis of the measured sample surface. For all tested materials, the measured surface is set to at least 10 × 10 periods of the expected waviness depending on the fabric structure (see Figure 6). A sensitivity analysis of the point density between 20 µm and 45 µm per measuring point on the organic sheets shows no significant influence on the maximum waviness. Therefore a grid of 45 points/mm is used for efficiency reasons. After the scan of the surface, the raw data is evaluated with the software FRT Mark III (version 3.9.13). Through the use of the boundary wavelength λc the primary data is divided into a waviness and a roughness range, respectively [23,24]. A further differentiation of the waviness to longer wavelength is not possible in the software. The calculated waviness is strongly dependent on the adjusted border wavelength λc. Therefore, the theoretical border wavelength λc is calculated from the periodicity of each fabric structure. The border wavelength is then set to 90% of the theoretically determined value to be able to measure small variations



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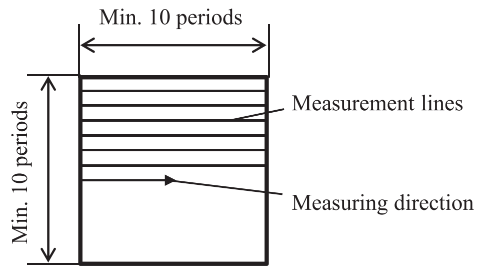

(b)

Figure 6: (a) Picture of the white light profilometer measurement head with sample specimen, (b) Schematic drawing of the measurement area and measuring direction.

in the fabric geometry. Due to the periodic structure of FRPC surfaces, the maximum waviness Wz and Wz25 are used in this work for the characterization of surface perception. Wz is defined as the arithmetic average of the height of profile between the height of the largest profile peaks and the depth of the largest profile valleys within the sampling length. Wz25 is defined as the arithmetic average of the height of profile between the height of the largest profile peaks and the depth of the largest profile valleys within a sampling area, which is divided into 5 × 5 equal fields, instead of a sampling length. This allows to reduce the impact of anisotropies, which may be present in the investigated laminate surfaces.

4 Results and Discussion

The influence of textile parameters fiber diameter d and mesh size w on the surface waviness will be examined. As shown in Figure 7 an increase in fiber diameter or mesh size increases the maximum

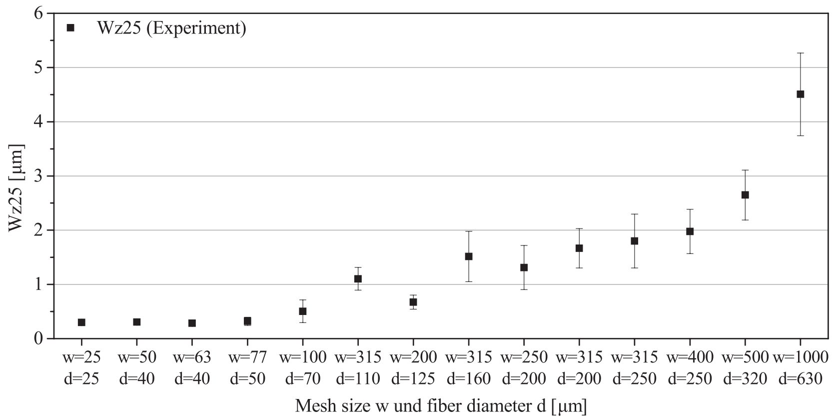

Figure 7: Maximum waviness Wz25 in dependence from fiber diameter d and mesh size w for 14 different steel wire fabrics.

waviness Wz25. For fiber diameters d between 25 µm and 50 µm there is hardly any variation measurable. Wz25 is around 0.3 µm for all 4 laminate configurations. This results from the maximum resolution of the used white light profilometer, which is about 0.3 µm. A periodic surface texture caused by the fabric below this limit amplitude can no longer be resolved by the measurement device.

For the roughest fabric (d = 630 µm), the maximum waviness is approximately 4.5 µm. For filament fabrics in technical applications, typical roving heights, which depend on the used fiber architecture, the yarn count and the fiber volume content are between 100 and 200 µm [25]. Steel wire fabrics with comparable fiber diameters achieve maximum waviness in the range of 1 to 2 µm.

4.1 *Influence of fiber diameter and mesh size*

To investigate the influence of mesh size w four different fabrics with a constant mesh size w = 315 µm and variable fiber diameter were examined. The fiber diameter varies from 110 µm to 250 µm in otherwise identical laminate build up. An increasing fiber diameter leads to an increased waviness. Under the assumption that the waviness forms due to the difference of the negative thermal expansion between fiber- and matrix-rich zone, a linear increase in waviness with increasing fiber diameter is expected (see eq. (4)). Including the standard deviations of the measured values, a linear dependence of the waviness from the fiber diameter can be found in the experimental data (see Figure 8).

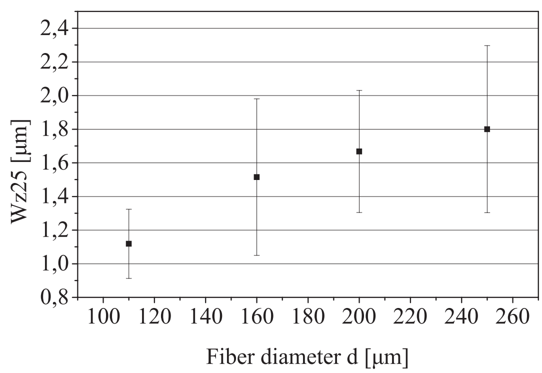

Figure 8: Maximum waviness Wz25 in dependency from fiber diameter at a constant w = 315 µm.

If the fiber diameter is kept constant and the influence of the mesh size on the maximum waviness is evaluated, waviness develops as shown in Figure 9. An increase in the mesh size moderately increases the maximum waviness. As the waviness is derived from the one-dimensional profile difference perpendicular to the surface it does not consider the volume shrinkages of fiber and matrix. An increasing mesh size increases the size of the shrinking volume and therefore increasing the waviness (see Figure 9).

An example with d = 250 µm illustrates the basic relationships. The theoretical thermal expansion calculated with eq. 4 keeps constant with varying mesh size due to the constant fiber cross section. If a fabric mesh is assumed to be cuboid, the mesh volume arises from a fabric with w = 315 µm, d = 250 µm as:

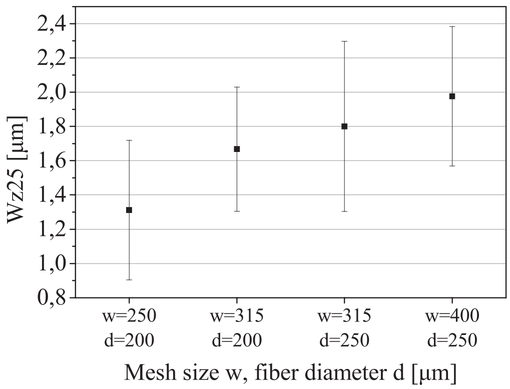

Figure 9: Maximum waviness Wz25 in dependency of the mesh size w at two constant fiber diameters.

$$V_0 = w^2 \cdot d = 315\,\mu m \cdot 315\,\mu m \cdot 250\,\mu m = 24.806.250\,\mu m^3 \tag{6}$$

and for a fabric with w = 400 µm, d = 250 µm:

$$V_0 = w^2 \cdot d = 400\,\mu m \cdot 400\,\mu m \cdot 250\,\mu m = 40.000.000\,\mu m^3 \tag{7}$$

The volumetric expansion derives from

$$\Delta V = V_0 \cdot 3\alpha_{th} \cdot \Delta T \tag{8}$$

An increase in mesh size from 315 µm to 400 µm leads to a 61% increase in volume shrinkage. However, the increase in maximum waviness for a fiber diameter d = 250 µm is 26.9% and for a fiber diameter of d = 200 is 8.3%. The lower increase in waviness is because the shrinkage occurs in all three spatial directions. This leads to complex thermo-mechanical interactions between fiber and matrix within the composite, resulting in shrinkage constraints.

The fabric parameters fiber diameter d and mesh size w have a significant influence on the resulting surface ripple. Both an increase in fiber diameter as well as an increase in mesh size leads to significantly higher waviness values. Organic sheets made of a fabric with a fiber diameter of d = 50 µm or smaller result in a minimal, measurable waviness. A further decrease in waviness cannot be detected by the measurement equipment and by the subjective human perception.

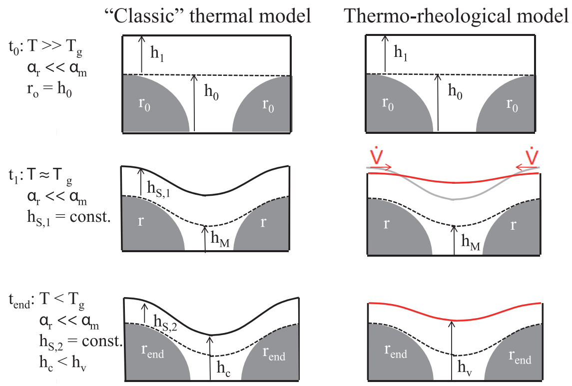

Figure 10: Comparison of surface development during processing between thermal and thermo-rheological model.

4.2 *Process model development - Analytic waviness correlation*

Considering the thermal shrinkage from chapter 3.2 the maximum waviness can be calculated. Figure 10 shows the theoretical waviness development under sole consideration of thermal shrinkage on the left side during thermoforming with rigid tools and sufficient long cooling times. Under the assumption that at the time t_0 (polymer is liquid; T = Tmax) the FRPC is ideally forming the molds flat surface, the cooling and thus shrinking of the FRPC starts directly after tool contact. The local differences in shrinkage cause the characteristic waviness, which, according to equation 4, is dependent on the height of the unit cell, the temperature- and pressure-dependent coefficient of thermal expansion α_{th}, as well as the maximum temperature difference of the organic sheet $\Delta T = (T_{OB}(t_0) - T_{OB}(t_{end}))$ during the processing. Because the material-specific properties (height of the unit cell, α_{th}) are largely process-independent, the temperature difference, as process-dependent factor, determines the absolute waviness Wz.

Figure 11 shows the theoretical calculated waviness values for the investigated organic sheet compositions. For the temperature-dependent CTE of polycarbonate the values given in table 1 and for the steel fibers a constant CTE $\alpha_r = 16, 5 \cdot 10^{-6}\ K^{-1}$ are used. The initial height is defined by the fabrics fiber diameter plus a 100 µm surface layer. The temperature difference ΔT is given by the temperature when cooling

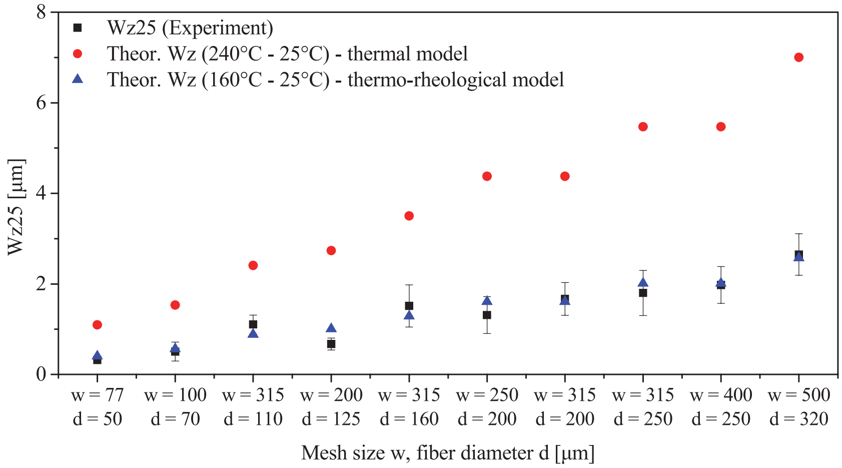

Figure 11: Waviness comparison between experimental results and analytical calculated waviness values for different fiber diameter and mesh size combinations.

starts (240°C) to room temperature RT. As can be seen, the calculated waviness for pure thermal shrinkage (simple model) leads to exaggerated waviness values. Therefore, the simple thermal model is not able to exactly describe the surface development.

To better understand the surface development during processing the rheological properties of the matrix have to be included. The significantly reduced viscosity of the matrix in the melted state influences the surface properties. Because the tool stiffness is several magnitudes above the stiffness of the liquid matrix, it is assumed that no tool deformation takes place during the process. In contrast, the incipient waviness generates local displacements of the matrix from the tool surface. This leads to surface deformations and local stress peaks in the matrix area arising in the contact area tool-FRPC. These inner tension concentrations are compensated by polymer flow processes of the low viscosity matrix from wave peaks to wave troughs. This partly compensates the thermally-induced waviness through a volume flow of the matrix. The matrix flow takes place as long as the so-called no-flow-temperature T_{NF} is reached. T_{NF} is specified as the temperature where externally applied tensions are less than the yield stress of the matrix and thus setting the threshold for polymer matrix flow. Upon further cooling, the classic thermal shrinkage dominates the surface development. By extending the thermal shrinkage to a thermo-rheological model the temperature difference ΔT is reduced. Starting temperature for thermally induced waviness is T_{NF} and end temperature is room temperature, which leads to lower waviness, compared to purely thermal shrinkage starting at 240°C.

To determine T_{NF} no standardized procedure or standard exists. Therefore, T_{NF} can be different for the same polymer depending on the method. In literature no-flow-temperature of polycarbonate is specified with a spectrum of 172 to 217°C [26].

For the recalculation of waviness including the rheological properties T_{NF} is set to 160°C. The slightly lower value compared to other publications results from the fact that the equation 4 does not consider any volumetric effects, which is corrected by reducing T_{NF}. As can be seen in Figure 11 the thermo-rheological model indicates a good correlation of the analytically calculated waviness values with the experimentally determined results.

5 Conclusions

This paper deals with the surface development of continuous fiber-reinforced thermoplastic composites, so called organic sheets, during variothermal processing. Organic sheets with different steel wire fabrics were manufactured using a variothermal laboratory press. The fabrics differed in fiber diameter and mesh size. Both textile parameters have a significant influence on the characteristic surface waviness of the FRPC. Increasing fiber diameters and mesh sizes lead to higher maximum waviness. Organic sheets with a fiber diameter less or equal 50 µm could not distinguished anymore and had no visible waviness. Furthermore, waviness was analytically calculated based on a model of thermal shrinkage. The so calculated values show no good correlation to the experimental values. Based on these results a thermo rheological process model was developed, which describes the waviness creation including a pressure induced compensating polymer flow during the variothermal processing. A waviness comparison of experimental data with model results shows a good accordance. The proposed model is able to predict surface waviness with input parameters being the laminate geometry, material CTEs as well as the rheological matrix behavior.

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