STUDY OF FIBER LENGTH AND MODELING OF PARTIALLY COMPACTED, COMMINGLED POLYPROPYLENE GLASS FIBER FLEECE COMPOSITES

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Abstract

In this work we investigated the influence of glass fiber content, number of layers and initial length on the residual fiber length and the properties of partially compacted composites made of commingled polypropylene and glass fibers. Furthermore, we wanted to develop a model to predict the properties of such composites.

We found, that despite of a significant degradation of fiber length due to the processing, increasing glass fiber content and initial fiber length leads to higher portions of longer fibers and mechanical properties are improved, but only when the porosity remains among certain levels. Porosity is therefore the critical factor influencing this type of composites. The modeling of the elastic modulus was found satisfactory for composites with porosity volume content under 0.5.

Introduction

Mechanical properties of composites can be significantly improved by the incorporation of reinforcing agents. Glass fiber reinforced composites have been widely used due to low cost, availability and very good mechanical properties. Among different types of glass reinforcements that can be used [1,2], the special kind of composite investigated in this study is produced from commingled fiber mats, i.e. swirl mats, where the reinforcement and the melt binder fiber is commingled and formed to a non-woven material which is then partially compacted in a hot press, which means that the final composite features certain porosity that is desired in order to achieve light-weight and other special properties such us acoustic insulation or ball impact properties.

Porosity was indeed found a very influencing factor for the final properties [3] of this kind of composite. Other factors such as final length of the glass fiber dispersed in the matrix are closely related to the final performance of composites and have been widely studied [4-6].

A simple model to predict composite properties is the rule of mixtures, as for example the approach by Kelly and Tyson [7], which is appropriate for fully compacted composites but not for partially consolidated composites, which feature porosities that influence final properties. The approach by Madsen and Lilholt [8] uses a factor $(1-V_p)^2$ to take the porosity into account, yielding the following equation:

$$E_c = (V_t E_t + (1-V_t)E_m)(1-V_p)^2$$  \hspace{1cm} (1)

which can be rewritten to:

$$\frac{E_c}{(1-V_p)^2} = V_t (E_tE_m) + E_m$$  \hspace{1cm} (2)

where $E_c$ is the ultimate modulus of the composite, $E_t$ and $E_m$ are the elastic modulus of the glass fibers and PP fibers respectively, $V_p$ is the volume fraction of the porosity and $V_t$ is the glass fiber volume fraction, which can be defined by Equation 3:

$$V_t = \frac{\frac{m_t}{\rho_t}}{\frac{m_t}{\rho_t} + \frac{m_m}{\rho_m} + V_p}$$  \hspace{1cm} (3)

where $m_t$ and $m_m$ are the mass fraction of the glass and PP fibers respectively, $\rho_t$ and $\rho_m$ their corresponding densities and $V_p$ is the volume fraction of the porosity.

The aim of this work was to investigate the influence of the glass fiber content, number of layers and initial length on the residual fiber length and thus on the properties of commingled, partially compacted composites. Furthermore, we wanted to find a model to predict the properties of such composites, i.e. the elastic modulus, to aid materials development.

Materials & Methods

Three series of fleeces were produced using 7 dtex (about 31 µm) polypropylene (PP) fibers with a staple length of 60 to 80 mm supplied by IFGAsota GmbH, in which 3% of a proprietary grade of maleic anhydride grafted polypropylene (MAH-PP) was used as additive. Glass fibers with an average fiber diameter of 17 µm, sized for the use in polypropylene, supplied by Owens Corning were used as reinforcement. Both materials were mixed by hand and processed into fleeces using a laboratory card.
150 g of the mixtures were introduced in the carding machine operating at a constant processing speed up to three times to achieve a better consistency and homogeneity of the mixtures. The fleeces with GSM values of about 500 g/m² were subsequently pressed between two plates in a hot press (Höfer H10) at 190°C and 30 bar for 15 minutes and then cooled down for 10 minutes at 20°C and 5 bar. The tooling used was a stamp forming tool, which produces plates with 100x100 mm².

Porosity was found critical for this kind of composite in our previous work [3]. In order to study the influence of residual fiber length on fleeces with different porosities two series were prepared: the first series was prepared using 0-50 wt% of glass fibers cut to a length of 160 mm and mixed with PP and pressed double-layered following the above-mentioned procedure. For the second series, the glass fiber content was fixed at 50 wt% and the number of layers was modified from 1 to 5 layers. The third series was prepared to investigate the effect of initial fiber length on the residual length and thus, the final properties of the composites. For these purpose the glass fibers were cut to a length of 80 to 200 mm in 40mm steps and mixed with an equal amount of PP fibers. In this case, 4 layers of fleeces were stacked, proved to be optimum number of layers to reach best mechanical properties [3].

Tensile properties were determined in accordance to ISO-527 in a Zwick Roell Z020 universal testing machine at a crosshead speed of 1 mm min⁻¹ for determining the elastic modulus and afterwards with 5 mm min⁻¹ until the break of the samples, with three replicates per series. For this purpose test samples with a size of 90x10 mm² were cut from the fleeces. The determination of porosity is critical for the correct interpretation of the final properties of the composite and it is decisive to carry out the measurements for each sample using the remaining material from the tensile tests for better correlation of results. First, the apparent density (ρ_{app}) was determined by weighing the specimen with known volume following Equation 4:

\[ \rho_{app} = \frac{m_c}{v_c} \]  

(4)

where \( m_c \) is the weight of the composite and \( v_c \) its volume, determined by mechanically measuring the dimensions.

Second, the density of the composite in a liquid capable of filling the porosities had to be measured. In this case ethanol was used as an immersion fluid and the density was measured according to ISO 1183 with a density-kit Sartorius YDK01 in combination with a Sartorius lab scale as shown in Equation 5:

\[ \rho_{EtOH} = \frac{m_{GF} + m_{PP}}{v_{GF} + v_{PP}} \]  

(5)

where \( m_{GF} \) and \( v_{GF} \) are the weight and volume of the glass fibers and \( m_{PP} \) and \( v_{PP} \) those of the PP fibers, respectively.

Finally the porosity of the composites (V_p) was calculated with the following equation:

\[ \rho_{EtOH} = 1 - \frac{\rho_{app}}{\rho_{EtOH}} \]  

(6)

2x2 cm² samples were cut from the composites and burned by heating them up to 625 °C using a Lecro Macro TGA 701 Machine in air atmosphere, in order to determine the final fiber weight content on the one hand and separate the glass fibers from the matrix on the other hand. The extracted fibers from the sample were separated manually. The residual glass fiber length was determined by image analysis. At least 1.500 fibers were measured using a high resolution scanner and FASEP software system. The number average fiber length (\( l_n \)) and the weight average fiber length (\( l_w \)) were obtained using the following equations [6]:

\[ l_n = \frac{\sum n_i l_i}{\sum n_i} \]  

(7)

\[ l_w = \frac{\sum n_i l_i^2}{\sum n_i l_i} \]  

(8)

Where \( l_i \) is the length of the i\(^{th}\) fiber in the sample and \( n_i \) is the sample frequency with the length increment range \( l_{i+1} - l_i \).

**Results & Discussion**

**Influence of glass fiber content**

As it can be seen in Table 1, the number average fiber length \( l_n \) is smaller than weight average \( l_w \), since the first is influenced by the amount of fibers and the latter by the fraction of long fibers present. Looking at the D values of the fiber length distribution (representing the midpoint and the range of particle size at 10% and 90% of fibers), the weight average correlates with the D_{50} value, and is thus more meaningful for the final properties of the composite, since these are influenced by the volume of fibers rather than the number of fibers.
Table 1: Glass fiber residual length parameters calculated from the length distribution; all values given in mm

<table>
<thead>
<tr>
<th>wt %</th>
<th>I_n</th>
<th>I_w</th>
<th>D_{10}</th>
<th>D_{50}</th>
<th>D_{90}</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.86</td>
<td>5.98</td>
<td>2.09</td>
<td>5.18</td>
<td>11.73</td>
</tr>
<tr>
<td>20</td>
<td>1.08</td>
<td>5.83</td>
<td>1.99</td>
<td>4.96</td>
<td>11.20</td>
</tr>
<tr>
<td>30</td>
<td>1.29</td>
<td>5.71</td>
<td>1.67</td>
<td>4.86</td>
<td>10.96</td>
</tr>
<tr>
<td>40</td>
<td>1.48</td>
<td>7.03</td>
<td>2.40</td>
<td>6.06</td>
<td>13.16</td>
</tr>
<tr>
<td>50</td>
<td>1.49</td>
<td>8.63</td>
<td>2.7</td>
<td>6.85</td>
<td>15.6</td>
</tr>
</tbody>
</table>

Residual fiber lengths show that there is a clear breakage of fibers during processing, since the initial fiber length of 160 mm decreases to a weight average between 5 and 9 mm. However, increasing fiber content leads to longer residual fibers as it can be seen in Figure 1, where the relationship between the residual fiber length and the glass fiber content is depicted. The processing with the carding machine has more influence on fiber length than fiber-fiber interactions in this case, thus resulting in higher portions of longer fibers if more glass fibers are incorporated.

The relationship between mechanical properties and weight average length is depicted in Figure 2, in which porosity and glass fiber content are printed beside the data points. As it can be observed from the data points given for 40 and 50 wt% of glass fibers, the porosity is the key factor influencing the mechanical properties, since the increase on fiber content and residual length do not lead to better properties due to the increase in void content.

Figure 2: Elastic modulus and tensile strength of the composites with different glass fiber content

**Influence of layers**

With the glass fiber content set at 50%, the variation of the number of layers shows an influence on the residual length, which decreases with increasing number of stacked layers (Figure 3). This might be attributed to the increase of fiber collision because the interaction between adjacent fibers is intensified, as well as fiber-matrix interaction during pressing as higher amount of PP- and glass fibers are available.

Figure 3: Fiber length distribution vs. number of layers for glass fiber PP composites

Figure 4 shows the elastic modulus and tensile strength of the composites in relation to the weight average of the residual length. Also in this case porosity proves to be decisive for the mechanical properties, whereas an increase in residual fiber length does not result in improved properties.
Influence of initial glass fiber length

Considering the results of the previous sections, the last question to be considered was if the residual fiber length could be critical in the case of comparable porosities. With this purpose, the initial glass fiber length was altered whereas the remaining parameters where kept constant. Similar porosities were yielded for all composites with values between 9-12%.

The residual fiber length increases with increasing initial fiber length (Figure 5). However, the achievable maximum length seems to be limited to approximately 3.6 mm. The number of layers and thus the intensified fiber-fiber interactions may be the reason for this limitation as it can be seen as well in Figure 3.

![Figure 4: Elastic modulus and tensile strength of the PP glass fiber composites with different number of layers](image)

![Figure 5: Residual fiber length vs. initial length for PP glass fiber composites](image)

Modeling approach

The results from the previous sections show that fiber length is not the key influencing factor regarding mechanical properties for this kind of composite, but porosity. For this reason, the porosity factor from Madsen and Lilholt [8] as in Equation 2 was applied for the experimental results of elastic modulus and plotted against the fiber volume content (Equation 3) calculated from the fiber mass contents determined by TGA analysis (Figure 7). As it can be observed, the two data points corresponding to the composites with the highest porosity values (57 and 73%) differ significantly from the rest of the data and show that it’s not possible to model properties of composites with such high porosities. However, by fitting the elastic modulus of the remaining data the suitability of the applied model is demonstrated.

![Figure 6: Elastic modulus and tensile strength of the composites with different initial fiber length for PP glass fiber composites](image)

The theoretical values of $E_m$ and $E_p$ can be obtained applying Equation 3 and equate to 1,956 and 28,578 MPa respectively. The elastic modulus of the glass fibers differ significantly from the values of 73 GPa found in literature [9]. However, applying the orientation factor for random in plane oriented fibers, thus accounting for the fiber deviation from the main direction of loading $n_1=0.375$ [10], one gets about 76 GPa. This is in excellent accordance to the literature data, especially taking into account that the carding operation does not produce completely random in-plane orientation, but has some higher orientation in the machine direction.
Conclusions

The influences of glass fiber content, layer number and initial length on the residual fiber length and the mechanical properties of partially compacted glass fiber-PP composites were studied. We found that there is a significant degradation of the fiber length mostly due to the processing with the carding machine but also due to intensified interactions between adjacent fibers as well as between fiber and matrix during pressing when the number of layers is increased and thus more matrix and fibers are available. Increasing glass fiber content and initial fiber length leads to higher portions of longer fibers, however the achievable maximum length is limited.

Mechanical properties are improved with increasing initial fiber length and thus residual fiber length in the composite as long as the porosity remains among certain levels, typically below 25%. No influence of fiber length could be observed when porosity levels increase. For comparable porosity values the increase on properties due to longer fiber length is limited as well, especially after the length weighted residual glass fiber length reaches approx. 3 mm. This can be seen as a supercritical fiber length for the investigated composites, and comparing this with data from injection molded glass fiber reinforced PP, we find – depending on the exact combination – similar critical fiber lengths between 1 and 2.5 mm [5].

As stated above, porosity is the controlling factor for the mechanical properties and for this reason was the critical factor for the modeling approach. Excellent agreement between experimental data and the model of Madsen and Lilholt [8] was obtained over the range of fiber lengths and contents for composites with porosity volume content under 0.5, thus providing a tool to predict composite properties.

References


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Key Words: glass fiber, polypropylene staple fiber, Rule-of-Mixtures, modelling