INFLUENCE OF PROCESS PARAMETERS ON POROSITY AND MECHANICAL PROPERTIES OF STRUCTURAL THERMOPLASTIC COMPOSITES MADE BY HOTPRESSING

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Abstract
For the characterization of structural plastic composites, layers of glass fibre fabrics and polyamide sheets were alternately stacked and pressed to consolidated composite under different processing pressures and temperatures. To investigate the influence of the fabric type in correlation with the process parameters, different types of glass fibre fabric were used. From the produced sheets test specimens were cut out for tensile-, bending- and Charpy-impact characterization. We found that with higher processing pressure the porosity content decreases while the impact- and bending strength stay fairly constant. In addition we found, that the twill weaved fabrics are easier penetrated by the thermoplastic matrix than plain weave which leads to better mechanical properties with comparable fibre volume fraction.

1. Introduction
Structural plastic composites (also referred to as organo sheets) have an excellent fracture toughness and thermoforming behaviour, low density and, above all, they are recyclable, which is of growing importance, as sustainability is often also a legal requirement, as shown through the End of Life Vehicles Directive of the European Union. One issue of thermoplastic matrix systems is the much higher viscosity which makes the production process complex and costly whereby industrial applications are still low but progressively growing. To facilitate that grow an enhancement of the process ability without sacrificing product performance is of great importance.

Therefore the aim of this work was to investigate the influence of the production parameters of the pressing process on the porosity and the structural and mechanical properties of structural plastic composites.

2. Materials & Methods
The materials used were different types of glass fibre fabric (table 1) and 0.5 mm thick polyamide sheets (MFR 40 g/min at 250 °C and 2.16 kg). The fibre and polymer sheets were cut, alternately stacked and pressed with a hydraulic heat- and cooling press, where different temperatures and pressures were applied. For the first test series an unmodified pressing tool made of aluminium was used. Later a new mould made of steel was developed to achieve a better sealing and avoid leakage of the matrix at higher pressures.
Table 1. The used glass fibre fabrics and their characteristics from the data sheets. \( m_a \) is the surface weight, \( \rho_f \) the fibre density.

<table>
<thead>
<tr>
<th>Fibre Type</th>
<th>( m_a ) [g/m²]</th>
<th>( \rho_f ) [g/cm³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>GF Twill 2/2</td>
<td>390</td>
<td>2.45...2.58</td>
</tr>
<tr>
<td>GF Plain</td>
<td>390</td>
<td></td>
</tr>
</tbody>
</table>

For each experiment 8 layers of PA sheets and 7 layers of fabric were stacked in the pressing mould and pre-heated for 20 minutes. After 2 minutes of applying the pressure, the tool was cooled down and the organo sheet was demoulded. To evaluate the influence of the process parameters the pressing temperature and pressure was changed in each experiment. From the so produced plates specimens for mechanical characterisation were milled out. Figure 1 shows an organo sheet with the milled out specimens for bending-, tensile-, impact- and thermo-gravimetric analysis.

Figure 1. Pressed organo sheet where specimens were milled out for mechanical and thermal analysis.

The fibre mass content was evaluated by means of thermo-gravimetric analysis according to ISO-1172 where specimens were weighed before and after ashing. From this value the fibre volume content was calculated using a glass fibre density of 2.58 g/cm³ and a polyamide density of 1.15 g/cm³. The porosity content \( \Delta V_f \), which is crucial for the mechanical properties of the composite, was calculated by comparing the theoretical fibre volume content with the results from the thermo-gravimetric analysis (1). For the calculation of the theoretical fibre volume content \( V_{f_{calc}} \) formula (2) was used.

\[
\Delta V_f = V_{f_{TG}} - V_{f_{calc}}
\]

\[
V_{f_{calc}} = \frac{\rho_c w_f}{\rho_f}
\]

Where \( w \) is the mass content, \( \rho \) the density and the indices are \( f \) for fibre, \( p \) for polyamide or \( c \) for composite. The density of the composite \( \rho_c \) can be measured by means of a scale with a density determination kit according to ISO-1183 or calculated using formula (3):

\[
\rho_c = \frac{1}{\left(\frac{w_f}{\rho_f} + \frac{w_p}{\rho_p}\right)}
\]

2
For the characterization of the mechanical performance of the produced materials, Charpy impact-, tensile- and bending tests were carried out. Furthermore experiments for interlaminar fracture toughness (Glc) were performed but could not be evaluated due to non-laminar failure mode for all the investigated samples. The results from the tensile tests are not yet reliable, but still under development as the tab bonding has to be improved. To further investigate the wetting of the fibres, embedded and polished cross-sections were analysed by means of an optical microscope.

3. Results & Discussion

The preliminary mechanical results show the influence of the processing parameters, whereby higher pressures lead to a higher fibre volume content and further to higher bending modulus (figure 2). However over 100 bar mould pressure this effect is reversed, because of an unadjusted tool whereby the amount of matrix material that can be pressed through the fabric is limited. This leads to a low fibre volume fraction which was about 25% and poor mechanical properties. Nevertheless it can be seen, that when equal processing parameters are applied but the used woven fabric type is plain instead of twill weave, the fibre volume fraction further decreases (figure 3).

As a consequence a new pressing tool with improved sealing was developed. As the preliminary results showed that 250 °C is the suitable process temperature, this was used for all further experiments. The results show that the influence of the processing pressure is not
limited. We found that pressures of 50 bar lead to the highest bending modulus (figure 4). We assume that this is due to the fact that with lower pressure the fibres are less compacted and therefore the fibres are less misaligned due to the matrix flowing through the fabrics. Furthermore figure 4 shows that all the structural plastic composites produced achieved good mechanical values compared to conventional glass fibre reinforced plastic with thermoset matrix.

![Figure 4](image)

**Figure 4.** Flexural modulus and bending strength versus processing pressure for composites produced in the improved tool. The coloured areas indicate the range of values of a comparable thermoset composite.

On the other hand side, the calculated porosity content is highest when only 50 bar pressure is applied in the production process. From this follows that porosity lower than 3.8 % is not influencing the flexural modulus. Figure 5 shows the fibre volume fraction assessed by thermal gravimetric analysis and from density measurements. The porosity content was calculated using formula (1). From these results can be followed, that the realised fibre volume fraction of more than 50 % is good for that manufacturing process, and the results are quite comparable due to the overall low porosity.

![Figure 5](image)

**Figure 5.** Fibre Volume Content ($V_f$) and porosity ($\Delta V_f$) versus processing pressure for composites produced in the improved tool.

The optical micrographs prove the low porosity content as there are no porosities visible which can be seen in figure 6. Furthermore the pictures show the good wetting of the fibre with the thermoplastic matrix. Nevertheless closer investigations with optical microscope and scanning electron microscope will be done to evaluate the fibre-matrix wetting.
One of the major benefits of thermoplastic composites is their high impact strength. Figure 7 shows the unnotched and notched impact strength whereby no significant dependency on the processing pressure could be seen. Nevertheless in this case the values for the 50 bar specimens are a little bit lower which could be due to the higher porosity content.

4. Conclusions

The investigations with an improved pressing tool showed that a porosity content below 4 % can be achieved. The fibre volume fraction of about 55 % is compatible with thermoset composites and so are the mechanical properties. The twill weaved fabrics are easier penetrated by the thermoplastic matrix than plain weave which leads to better mechanical properties with comparable fibre volume fraction. Furthermore we found that the processing pressure has no big influence concerning the bending stiffness and impact strength. The flexural modulus is even higher at 50 bar pressure, which leads to the conclusion that the pressure on the fibres and in succession the misalignment reduces their bending strength. We found that Charpy impact strength is very high for the tested organic sheets, especially the values from the notched samples.

In general our work shows, that mechanical analysis like tensile-, bending- and Charpy impact-test are applicable and the performance is near thermoset composites with comparable fibre volume content. For the tensile tests the tab bonding is an issue which has to be improved. Further investigation will be done with different types of fabric and fibre materials as well as recycling of the organic sheet which is another big benefit of the thermoplastic composites.