INFLUENCING FACTORS ON THE PROPERTIES OF NATURAL FIBER REINFORCED THERMOPLASTICS PROCESSED BY INJECTION MOULDING

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SUMMARY: Injection molding is a suitable method for processing natural fiber reinforced composite parts. The most important factors influencing the resulting mechanical properties are the quality of the fiber matrix adhesion, the selection of suitable processing parameters and the fiber orientation. It is possible to separate the fibers more efficiently and to increase the specific surface by means of appropriate decortication procedures and alkali treatment. This leads to a higher reinforcement potential of the natural fibers. With the help of thermal analysis it is possible to characterize the limits of the processing temperatures for natural fibers as well as for the thermoplastic matrix. In order to take into account the low temperature resistance of natural fibers suitable processing parameters need to be chosen. The matrix flow during the injection molding process results in a layer morphology and causes anisotropic mechanical properties.

KEYWORDS: Natural fiber reinforcement, treatment of natural fibers, thermoplastics, injection molding, layer morphology, thermal analysis.

INTRODUCTION

Flax and hemp are renewable resources which provide biodegradability and CO2-neutrality. Composites of natural fibers and thermoplastic matrices combine good mechanical properties with a low specific mass [1]. Flax (on account of its strength) and hemp (because of its high amount of straw and fibers) are presently predominantly applied in Germany. Suitable matrix materials for natural fiber reinforced polymers are resin systems, thermoplastic starch and polyolefins (polypropylene and polyethylene), while polypropylene provides most of the advantages with regard to economic (price), ecological (recycling behavior) and technical requirements (higher thermal stability than polyethylene).

Injection molding is a very suitable procedure to process natural fiber reinforced polymers into sophisticated 3-dimensional parts. In this case, the use of granular material, which already includes fibers and coupling agent, proves a success. The compounding process takes place on twin screw extruders because of its high distributive and dispersive mixing efficiency.
FIBER MATRIX ADHESION

Fiber matrix adhesion of fiber composite parts is influenced by various factors. Essentially, these are:

- Specific surface of reinforcing component
- Compatibility of components
- Processing parameters (pressure, temperature)

Fiber treatment

Flax and hemp fibers can be mercerized boiling in a 10 % soda solution as well as in a 2 % sodium hydroxide solution. Unwanted fiber ingredients dissolve during the boiling process. After repeated washing with distilled water and the following drying, it is possible to discover changes with respect to the basic material.

- Hurds can be removed more easily,
- Fibers are purified,
- Unwanted fiber ingredients are largely removed,
- Fiber separation ability is increased.

Fig. 1: Untreated flax fibers  Fig. 2: Mercerized flax fibers

SEM-pictures explicitly reveal the success of the treatment. There is no conversion from cellulose I to cellulose II under the chosen treatment parameters. The enlarged specific surface is shown when comparing figure 1 with figure 2.

Specific surface

The fiber matrix adhesion is dependent, among others, on the specific surface of the natural fibers. Due to the special fiber separation processes the specific surface can be enlarged to modify the interface between fiber and matrix.

The measurement of specific surface and porosity by means of the BET-method (Brunauer, Emmett, Teller) works on the basis of physisorption, i.e. the adsorption and desorption of gases (mostly nitrogen) on the outer and inner surfaces of a substance. Owing to the physical conditions, natural fibers correspond to type II out of five classified isothermal characteristics according de Boer [2]. Because of the very small specific surfaces it is not possible to apply the 1-point-method. That is why the measurements have been executed with the gas
adsorption device Sorptomatic 1990 using a special measuring vessel. Figure 3 reveals the resulting specific surfaces.

![Specific Surface of Natural Fibers](image)

**Fig. 3: Specific surface of different natural fibers**

Flax fibers have a specific surface approximately three times as big as the specific surface of hemp fibers. Steam explosion separated flax occupies a special position. With the help of this particular treatment procedure, it is possible to achieve a maximum special surface.

The influence of the specific surface on the macroscopic properties of the component parts is mirrored, among other things, in the mechanical properties of the composite. Using 30 wt-% retted purified flax, washing with alkali increases tensile strength from 39.5 MPa to 45.4 MPa. Simultaneously, the characterization of the specific surface is an indication of the successful removal of fiber companions.

**PROCESSING PARAMETERS**

Natural and synthetic polymers are subject to a degradation of the mechanical properties under the influence of increased temperatures [3]. The thermal stability of the natural fibers and the polypropylene matrix that are to be processed is characterized by means of thermogravimetric (TGA) and differential scanning calorimetric analysis (DSC).

Exceeding the restricted processing parameters causes oxidative degradation and reduction of the polymer chain length. Weight loss occurs up to approx. 140 °C because of the vaporization of adsorbed water. Exposed to air atmosphere, the mass of natural fibers continuous to decrease slightly between 200 and 220 °C, above this temperature, an irreversible degradation of the fibers occurs. The decrease of the mechanical properties of thermal treated fibers was investigated by single fiber tensile test. A reduction of the processing duration will in any case have favorable influence on fiber and matrix properties.
The melting range of the polymer matrix is visible as an endothermic peak in DSC. The material used shows an onset value at 159 °C and a specific melting heat of 98 J/g. The exothermic peak in the temperature range of about 200 °C can be traced back to the history of the polypropylene. The repeated measurement does not show an exothermic reaction. The measured melting temperature of the matrix is the least possible processing temperature.

![DSC of Polypropylene (N₂), TGA of Flax (Air)](image)

**Fig. 4:** DSC of polypropylene in N₂ and TGA of flax in air atmosphere

By means of well chosen processing parameters, in particular temperature and pressure, compounds with good mechanical properties are created. Table 1 shows suitable processing parameters for natural fiber reinforced polypropylene.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Melt temperature</td>
<td>200 °C</td>
</tr>
<tr>
<td>Mold temperature</td>
<td>50 °C</td>
</tr>
<tr>
<td>Volumetric flow rate</td>
<td>30 cm³/s</td>
</tr>
<tr>
<td>Holding pressure</td>
<td>60 MPa</td>
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**Table 1:** Processing parameters during injection molding

**LAYER MORPHOLOGY**

The achieved mechanical properties are mainly influenced by the material structure. A layered structure is typical for short fiber reinforced injection molded parts [4]. Some out-of-plane orientations of fibers can be usually neglected [5].

In order to be able to investigate the morphology of an injection molded rectangular plate in the transmitted light microscope, thin cut sections (15 µm) from the middle of the plate have been prepared. The cuts have been executed parallel to the surface. Figure 5 shows the
Fig. 5: Layer morphology
location of the thin cut sections and the transmitted-light photomicrographs. Similarly to what is known from glass fiber reinforced thermoplastics in the surface layer ($z = 0.9 \text{ mm}$), the main fiber direction is parallel to the flow direction, whereas in the following transition region ($z = 0.6 \text{ mm}$) fiber orientation is nearly random. In the core ($z = 0$) there is a thin layer with the main fiber direction normal to the flow direction.

This layered structure causes anisotropic mechanical properties [6] which will be characterized by means of suitable testing methods. Modulus in flexure and modulus in tension are differently influenced by the fiber orientation. Figure 6 shows the anistropy (modulus in flow direction divided by modulus in transverse flow direction) of specimens which were cut from the plate. The reason for the stronger anisotropy in the bending test lies in the considerable influence of the orientation of the skin layer.

![Figure 6: Anisotropy of injection molded plate](image)
CONCLUSIONS

The quality of the fiber matrix adhesion of natural fiber reinforced thermoplastics is influenced by fiber decortication and fiber treatment. In order to achieve maximum mechanical composite properties, the natural fibers have to be separated to a high degree and to be treated with alkali. The thermoanalytic investigations demonstrate that a temperature range between 190 °C and 220 °C has to be maintained in order to process flax fiber reinforced polypropylene successfully. Flow induced fiber orientation causes a layer morphology. The resulting anisotropic properties have to be considered when designing the requested parts.

Taking into account the above mentioned influences, composites can be manufactured successfully using injection molding. They represent a suitable material which is an alternative to glass fiber reinforcements many of applications in the field of lower mechanical loads.

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REFERENCES


