## Abstract

The work presented here supports the industrial use of natural fibre reinforced composite materials under mass production circumstances. Potentials for optimising the materials' properties are offered and evaluated with regard to their effect on the process chain material – coupling agent – processing. The possibility to use these materials in mass production applications are improved by optimising each partial stage.

Throughout the world there exists a great variety of suitable applications for this group of composites affecting the raw materials choice. The Europe's market is stamped by the requirements of the automotive industry, the important markets of Japan and the USA are dominated by civil engineering and landscaping applications. A yearly increase of 18 % in Europe, 25 % in Japan and 14 % in the US is expected. The US market offers the largest market volume of more than 480000 t exceeding the European Market for nearly five times.

To enhance the fields of application for natural fibre reinforced thermoplastics the common techniques of the film-stacking and the compression moulding process are used to manufacture optimised composites based on polypropylene and bast (hemp, flax) as well as leaf (sisal) and wood (spruce) fibres. Therefore new semi finished parts for the compression moulding process had to be developed.

Within the manufacturing of natural fibre reinforced polypropylene using the film-stacking process material and process parameters were identified to transfer the gathered knowledge to the compression moulding process. It has been seen that most problems are caused by the organic origin of the fibres. Especially the addiction of the fibres to decompose when treated with higher temperatures under pressure hampers their use in thermoplastic composites.

By investigating wood fibres as reinforcements, which differ from bast fibres in their chemical composition the influence of the process parameters temperature and pressure on the composite properties were evaluated and verified for hemp fibre reinforced polypropylene. The minimum process time was observed and in order to enhance the fibre-matrix-adhesion by using coupling agents the diffusion of the coupling agent molecules was determined theoretically. Therefore a model was evalu-

ated dealing with the maximum mass flow of coupling agent being transferred in the fibre-matrix-interface because of mass transfer mechanisms.

In order to optimise the wetting of the fibres with the matrix different possibilities to modify the fibres were investigated. Drying the fibres prior to the manufacturing of the composite is an easy and effective way to improve the fibre-matrix-adhesion. The tensile strength of all composites rose conspicuously. The removal of dust and water soluble substances by washing led to a higher tensile strength only with the sisal fibre reinforced composite. Washing the other fibres led to decreasing fibre wetting.

Fibre substances like lignin and pectin were removed using the mercerisation technique. Composites made from these chemically retted fibres show the more disintegrated fibre structure and a worse wet ability of the fibre surface with the polypropylene. Hence the tensile and bending strength was not enhanced. The Charpy impact strength of the composite raised distinctly.

The use of coupling agents based on maleic acid crafted polypropylene led to an increasing tensile strength up to 58 % compared to the composite manufactured with pre-dried fibres. The bending strength raised about 109 %. The Charpy decreased about 60 to 80 %. Flax fibre reinforced composites showed the highest tensile strength, sisal fibre reinforced composites offered the highest Charpy. No differences between copolymeric and homopolymeric polypropylene when using PP-MAH as coupling agents were determined.

The kind of application of the coupling agent in the compound has an major effect on the amount of coupling agent to be added. The closer the coupling agent is brought to the fibres surface at the beginning of the impregnation step the less amount has to be used. If using an aqueous suspension the least amount had to be added as the coupling agent remains directly on the fibre surface after drying. Mixing the coupling agent with the polypropylene hinders the well dispersed PP-MAH to act in the fibre-matrix-interface effectively, so the amount of coupling agent has to be increased.

The comparison of coupling agent containing compounds which differ in the amount of coupling agent and the molar mass distribution showed the amount of coupling agent related to the mass of fibres to be the important parameter to dose the PP-MAH. The mean molar mass distribution had no effect on the compounds' properties.

Transferring the knowledge gained from the film-stacked composites to the compression moulding process offered the possibility to use jute long fibre reinforced granules (LFT) under optimised processing conditions. The composites gained from the molten and pressed granules showed the highly dependency of the mechanical properties to the fibre direction in the part. If the fibres are able to flow along the cavity and direct themselves into parallelism the tensile and bending strength increases in the main flow direction and decreases perpendicular to this direction. The impact strength decreases with raising orientation of the fibres. The jute fibre surface presents a better adhesion to the polypropylene as the surfaces of the hemp, flax and sisal fibres, which could be improved by adding PP-MAH as coupling agent.

A newly developed pelletised semi finished part with sisal fibre reinforcement and the development of a direct impregnation process using solely a horizontal plasticating unit completes the work. Using an established plasticating extruder offers the possibility for the compression moulding industry to process natural fibre reinforced polypropylene with less investment. The compression moulded sisal fibre reinforced polypropylene showed varying fibre directions and disproportionate fibre-matrix-adhesion. As a result of the plasticating process in some parts bended fibres are still visible after compression moulding. Hence the used single-screw plasticator is not able to equalize the molten material. Increasing the compaction pressure was not possible as some parts showed beginning fibre degradation. Adding PP-MAH improved the fibre-matrix-adhesion but the positive effect of the materials' strength was not as clear as found for the film-stacked composites. Regarding the additional expenditures for compounding and the coupling agent costs the use of PP-MAH in compression moulded parts seems not to be useful.

Compared to the compression moulded glass fibre reinforced polypropylene from GMT and LFT-materials the natural fibre reinforced composites cannot reach the high level of material properties. Optimising the fibre-matrix-interface increases the properties but they are still lower than the properties of the glass fibre reinforced composites. Therefore the natural fibre reinforced materials are not able to substitute the traditional GMT and LFT, they rather should be used in new applications with lower demands.