**Introduction**

For high volume manufacturing, thermoplastic composites have proven to be very economic. This basically results from short cycle times and a high level of automation during processing. In 2004 the collaborative research centre CRC 639 was founded at TU Dresden to investigate such materials, especially hybrid yarn based thermoplastic composites. It is a collaboration comprising polymer science, textile technology, applied mechanics and process analysis, lightweight engineering as well as microelectronics. The central objective is to use textile processes to manufacture unique fabrics and textile preforms which can be processed in automated manufacturing cycles. The manufacturing path from the individual fibre to the composite structure is displayed in Fig. 1. Firstly, filaments have to be spun from the polymer melt and to be combined with glass filaments in a hybrid yarn. Subsequently, the yarns are processed by different textile technologies into fabrics and later on to near net shaped preforms. In order to manufacture a composite structure, the soft and “dry” textile preform has to be consolidated. After cutting, the structure can be assembled by several joining techniques [1].

The aim of the investigations is to provide adapted technologies for application-ready lightweight structures. This includes the consistent theoretical and technological description of the complete process chain from the individual filament to complex structures. As all steps in the process chain are highly complex, one exemplary material configuration is selected.

Hybrid yarns, consisting of polypropylene (PP) filaments and glass filaments (GF), are chosen as a base material for continuous filament reinforced thermoplastic composites. These yarns can be manufactured into adapted preforms with textile fabrication methods [2, 3]. They can be consolidated in short time pressing processes to structural parts with complex geometries and a high load-bearing capability [4].

It is well known that the mechanical properties of fibre-reinforced composites are influenced by the surface treatment (sizing) of the reinforcing fibres [5, 6]. The average thickness of these sizings’ is in the range of 0.5 to 1.0 µm for commercially available glass fibres with diameters between 10 and 14 µm. Generally, the sizing’s consist of 80-90 % by weight film former, 5-10 % by weight silane coupling agents and 5-10 % by weight auxiliary agents [7]. The functions of the silane coupling agents and their interactions with both the glass fibres and the polymer have been reviewed by
Ishida [8]. The generated silanol groups react with the glass surface primarily via hydrogen bonds to form partly covalent bonds. The second functionality of the sizing is the ability to interact with the thermoplastic or thermosetting resin by a chemical reaction. In earlier publications, it was shown that the interfacial shear strength can be improved by the application of modified polypropylene in short and continuous fibre-reinforced polypropylenes [9].

It was proven that the basic amino groups of the silane reacted with both the acidic groups of the PP matrix and the PP film former dispersion to build a strong covalent bond.

Another fact is that not only the silane coupling agent is important for interphase formation. Because of the extreme sensitivity to abrasion and bending of the reinforcing fibres, the sizing must also fulfil processing requirements, whereby it can influence the frictional behaviour. Moreover, it has been demonstrated that the film former plays a crucial role in interphase formation as reported for different model sizings [10]. Experimental results showed that the fibre-matrix interaction, tested by single fibre pull-out tests, depends on the chemical modification of the PP matrix in conjunction with the sizing. Highest interfacial shear strengths were determined with PP film formers and PP matrix, both modified by maleic-anhydride grafted PPs [10-13].

Due to matrix functionalization’s and their influence on the material behaviour of the composite it is necessary to evaluate and adapt existing material models. Thereby especially the behaviour of the thermoplastic matrix, affected by creep and relaxation phenomena has to be considered for accurate results. This also concerns the material behaviour under compressive loading in out of plane direction, like caused by clamping forces of screwed joints [14].

In this paper the inelastic material behaviour of modified unreinforced and textile reinforced PP has been investigated experimentally. A material model for polymers [15] was adapted to describe the material behaviour of the modified polypropylene and compared to the results of the experiments. Then a two-step homogenization procedure was used to predict the effective material behaviour of the composite.

Experimental investigation of modified polypropylene

The first part of the investigation of the behaviour of the modified textile reinforced PP deals with the calibration of the material model for PP using experimentally obtained data. For these experiments specimens were manufactured.

The homo-PP (HG455 FB, Borealis) with the average molecular weight \( M_w = 16 \times 10^4 \text{ g/mol} \) was used and blended with 2 % by weight of a maleic anhydride modified PP (Melt Flow Rate 36 g/10 min acc. to ISO 1133) which was produced from isotactic homopolymer by grafting with maleic anhydride (Exxelor P1020, Exxon Mobil Corp.) with \( M_w = 8.6 \times 10^4 \text{ g/mol} \) was used as base matrix material.

The specimens of modified PP (Fig. 2) were manufactured by injection moulding. The tension test specimens (160\( \times \)10\( \times \)4 mm\(^3\)), according to DIN 53455, specimen No. 3, (ISO 527.2) and plates (70\( \times \)80\( \times \)15 mm\(^3\)) were manufactured using the injection moulding machine Ergotech 100 (Demag Ergotech Wiehe GmbH). From these plates compression test specimen with the dimensions of 10\( \times \)10\( \times \)20 mm\(^3\) (length \( \times \) width \( \times \) height) were produced by milling.

![Fig. 2. Polypropylene specimens](image)

All tests include a set of five samples and were conducted at 23°C and 50% relative humidity. For the tensile tests a standard testing machine zwicki-Line Z2.5 (Zwick GmbH & Co. KG) with a video extensometer videoXtens® for the measurement of the elongation of the specimens was used. At the beginning, static tests with velocities of 0.5, 5 and 50 mm/min were conducted to determine the maximum stress level of the specimens. Then a relaxation test with a strain of 0.8 %, applied with a test velocity of 100 mm/min and a holding time of 1 h has been carried out. A creep test with a stress level of 15 MPa, applied with a test velocity of
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3.5 mm/min and the same holding time of 1 h is the last tensile test.

For the compression tests another testing machine Zwick UPM 1472 (Zwick GmbH & Co. KG) with parallel plates was used. The elongation was measured with the machine integrated measurement system. To avoid measurement errors due to the unknown elasticity of the machine, a compensation curve was recorded without a specimen before the beginning of the tests and used to calibrate the measured elongation of the specimen during the compressive tests. In accordance to the tension test, first static experiments were conducted with test velocities of 1, 10 and 100 mm/min. Afterwards, a relaxation test with 1.8 % strain, applied with a test velocity of 10 mm/min and a holding time of 1 h was performed. The final test was a creep test with a stress level of 20 MPa, applied with a velocity of 10 mm/min and the same holding time of 1 h.

The results of the static tests are shown in Fig. 4 and Fig. 5 (tensile tests) and Fig. 12 and Fig. 13 (compression tests). The first character in the caption of the diagrams is “T” or “C” for tension test or compression test. The second is “M” or “C” for matrix or composite and the third is “R” or “C” for relaxation or creep test in the long term diagrams. From Fig. 5 it can be seen, that the modulus as well as the stress level increase with higher test velocities. The same behaviour can be recognized in Fig. 12 for the compression tests of the matrix. Therefore, the material behaviour can be characterized as strain rate dependent which is a typical feature for polymers.

Calibration of the material model for the polypropylene

A small strain, isotropic viscoplastic material model proposed by Kästner et al. [15, 16] for modeling the inelastic constitutive behaviour of polymers is applied to represent the PP matrix in a numerical model of the local material structure of the composite. The constitutive relations are based on an overstress formulation, i.e. the total stress

\[
\sigma = \sigma^{eq} + \sigma^{ov}
\]  

is additively decomposed into the strain rate independent equilibrium stress \(\sigma^{eq}\) and the strain rate dependent overstress \(\sigma^{ov}\). In the case of present inelastic, e.g. plastic or viscous, deformations, the total strain

\[
\varepsilon = \varepsilon^{e} + q^{(i)}
\]  

is split into an elastic and an inelastic part which typically serves as an internal variable. The superscript \(( \quad ) \) will later relate this variable to a certain branch of the material model. Similar approaches are for instance followed by Haupt and Lion [17, 18], Hartmann [19] as well as Müller et al. [20, 21]. The structure of the used material model is illustrated by the rheological model shown in Fig. 3.

![Rheological model consisting of three principal model branches: a nonlinear spring, and endochronic model of plasticity and a set of \(\eta_{v}\) viscoelastic Maxwell models.](image)

The model includes a nonlinear elastic spring, an endochronic model of plasticity and a set of \(\eta_{v}\) viscoelastic Maxwell models.

The model of the equilibrium stress

\[
\sigma^{eq} = \sigma^{e} + \sigma^{end}
\]  

is defined by the combination of a nonlinear elastic stress-strain relation
\[ \sigma^e = \frac{E_1}{1 + \alpha |\varepsilon|} \varepsilon \]  
(4)

with material parameters \( E_1 \), \( \alpha \) and an endochronic model given by

\[ \begin{align*}
\sigma^{\text{end}} &= E_2 (\varepsilon - q^{\text{end}}) \\
\dot{q}^{\text{end}} &= \beta (\varepsilon - q^{\text{end}}) |\dot{\varepsilon}| 
\end{align*} \]  
(5)

and material properties \( E_2 \), \( \beta \). In this simple plasticity model, a hysteresis of the stress-strain curve for combined loading and unloading processes is represented by the internal variable \( q^{\text{end}} \) which can be interpreted as a plastic strain. Together these two model branches account for the strain rate independent parts of the material behaviour.

The constitutive formulation of the rate dependent fraction of the stress is a generalized Maxwell model with \( n_v \) parallel elements. The non-equilibrium overstress is the sum of the individual stresses in each Maxwell element

\[ \begin{align*}
\sigma^{\text{ov}} &= \sum_{i=1}^{n_v} \sigma_i^{\text{ov}} \\
\sigma_i^{\text{ov}} &= c_i (\varepsilon - q_i^{\text{ov}}) \\
\dot{q}_i^{\text{ov}} &= \frac{1}{\tau_i} (\varepsilon - q_i^{\text{ov}}) = \frac{c_i}{\eta_i} (\varepsilon - q_i^{\text{ov}}). 
\end{align*} \]  
(6)

In this model branch the internal variables \( q_i^{\text{ov}} \) are used to account for all rate dependent effects of the material behaviour, for instance the strain rate dependence observed in tensile tests performed at different velocities but also for the long-term relaxation and creep effects. The relaxation strengths \( c_i \) and relaxation times \( \tau_i \) (or equivalently the viscosity \( \eta_i \)) of each Maxwell element form a discrete relaxation spectrum whereat the number of required Maxwell elements is determined by the period of time which has to be modelled. Details on the required experimental stress-time data can be found in [22]. Typically one Maxwell element per decade of the covered time scale is used.

In [15] it was shown, that the stress-time curves obtained from tensile tests cannot be accurately reproduced using the linear overstress model, we therefore use a nonlinear strain rate dependence by replacing the constant viscosity \( \eta_i \) by a function

\[ \tilde{\eta}_i = \eta_i \exp \left( - \frac{|\sigma^{\text{ov}}|^{k_0}}{s_0} \right) \]  
(7)

of the overstress and two additional parameters \( s_0 \) and \( k_0 \). The idea of an overstress dependent viscosity function has previously been used for modelling of metals [17] and elastomers [18].

The procedure for the identification of the material parameters is in-line with the structure of the material model. Relaxation tests are an ideal means to quantify the parameters of the generalized Maxwell model, since they allow for a separation of strain rate dependent and independent fractions of the total stress. Therefore, the discrete relaxation spectrum is determined from a relaxation experiment using the window algorithm of Emri and Tschoegl [23]. All other parameters are obtained from fitting the model predictions to the results from tensile tests whereat information from [15] regarding the material behaviour of unmodified PP has been taken into account.

In Fig. 4 and Fig. 5 experimental and numerical results are compared. The simulation has been carried out with a finite element implementation of the generalized three-dimensional version [15, 16] of the presented material model. The comparison shows the good approximation of the rate dependent deformation behaviour in tensile tests (Fig. 4).

![Fig. 4. Comparison of results static tensile tests and simulation of the behaviour](image-url)
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POLYPROPYLENE

relaxation and creep experiments (Fig. 5). While the former experiments were used for the identification of material parameters, the latter comparison demonstrates the predictive capabilities of the model.

Experimental investigation of the composite

The second part of the investigation deals with the composite material. The first step is also the manufacturing of specimens. The process is shown in Fig. 6. From a multi-layered weft-knitted fabric made from a hybrid yarn containing GF and the modified PP square plates of 296×296 mm² were cut so, that the rovings in warp and weft direction are parallel to the edges of the square. These dimensions are a little bit smaller than the dimensions of the mould, so the preforms fit safely into it. Ten of these plates were laid up (Fig. 6 a) and consolidated in an evacuated mould in a laboratory press (300 kN pressing force, 200 °C mould temperature, 35 min holding time) to a plate with the dimensions of 297×297×6 mm³. Afterwards, stripes with a width of 20 mm and a length of 270 mm were cut out by water jet (Fig. 6 b). Four of these stripes were stacked in a second mould and pressed in an autoclave (60 MPa autoclave pressure, 200 °C temperature, 2 h holding time) to a bar of 20×23.6×270 mm³ (Fig. 6 c). In a last step specimen for compression test were made by milling from this bar (Fig. 6 d).

For the compression tests the same testing machine as used for the polymer, Zwick UPM 1472 (Zwick GmbH & Co. KG) with parallel plates, was utilized. To avoid measurement errors due to the unknown elasticity of the machine, the same compensation curve as in the compression tests of unreinforced PP was applied to calibrate the measured elongation of the specimen during the tests. First, static compression tests were conducted with test velocities of 1, 10 and 100 mm/min. Afterwards a relaxation test with 4 % strain, applied with a test velocity of 10 mm/min and a holding time of 1 h has been carried out. Finally, a creep test with a stress level of 117 MPa, applied with a velocity of 10 mm/min and a holding time of 1 h has been conducted.

The results of the static tests of the composite are shown in Fig. 12 and Fig. 13, with the same indices as in the previous diagrams. From Fig. 12 it can be seen that the composite clearly exhibits strain-dependent material behaviour. However, it is less pronounced than for the pure matrix material. One reason can be seen in the stiffening influence by the textile reinforcement. In comparison to the unreinforced specimens the stresses of the composite ones are about three times higher at the same strain level. The comparison of the plots of the long-time behaviour in Fig. 13 shows nearly the same characteristic but different stress levels between pure matrix and the composite.
Simulation of the effective mechanical response of the composite

In order to reproduce the experiments in numerical simulations, the material behaviour of all constituents and their geometrical arrangement in the composite have to be modelled properly. As a consequence of the hierarchical material structure, two consecutive homogenisation steps are performed to predict the effective behaviour of the composite according to Fig. 7. The first step is applied to areas of high fibre volume fraction which are replaced by a Homogeneous Substitution Material (HSM) called roving. The arrangement of roving’s of a certain textile reinforcement is the major characteristic of a mesoscopic Representative Volume Element (RVE).

Fig. 7. Two-step homogenisation procedure to predict the effective material behaviour of the composite.

The model for the PP matrix has been introduced in the previous section. Regarding the compressive tests, the long term behaviour is of particular interest. This can be reproduced using only a linear viscoelastic sub model of the presented constitutive relations consisting of a Hookean spring ($G_{oo}$) to account for the nonzero equilibrium relation and a set of $n_v$ Maxwell elements with modified parameters $G_j$ and $\tau_j$ for the strain rate dependent overstress. In the time domain a closed form, three-dimensional description of the model originally defined by equations (4) and (6) is given by the tensorial relaxation function

$$G(t) = G_{oo} + \sum_{j=1}^{n_v} G_j e^{-\frac{t}{\tau_j}}. \quad (8)$$

The equivalence of the full and the reduced linear sub model version for time intervals larger than $10^3$ s is shown by means of a numerical relaxation simulation in Fig. 8.

![Fig. 8. Comparison of the long term behaviour; numerical relaxation simulation](image)

As illustrated in Fig. 7, the roving itself consists of polypropylene and glass. Due to the PP matrix material, the roving will exhibit strain rate dependent behaviour. Hence, a homogenisation procedure is used to predict the effective, viscoelastic material behaviour of the roving. The microscopic RVE used for the homogenisation of the roving consists of the PP matrix material and glass filaments with a fibre volume fraction of 50% and the parameters $E_P = 73$ GPa and $\nu_P = 0.25$ [16, 24].

The used computational homogenisation approach is based on Hill’s principle, namely the equivalence of the averaged micro- and macroscopic stress power [25-27], in conjunction with periodic boundary conditions and the elastic-viscoelastic correspondence principle in combination with a Laplace-Carson transformation (LCT). Further details can be found in publications by Pierard [28, 29] and Haasemann et al. [30]. The associated algorithm is illustrated in Fig. 9.

![Fig. 9. Homogenisation based on elastic-viscoelastic correspondence principle](image)
For the LCT it is necessary to discretize the Laplace variable $s$ (see Fig. 9). A set of $n_{CT} = 8$ pairs of material tensors is used in the Laplace domain and for each a linear elastic homogenisation problem is solved. After the inverse LCT one obtains the effective, anisotropic viscoelastic material tensor $\tilde{G}(t)$. When the functions of the viscoelastic relaxation tensors in the time domain are known, it is possible to obtain the effective, anisotropic relaxation tensors for the HSM.

The results for two tensile loads applied transversely to the fibre as well as under an angle of 45 degrees ($T_2$, $T_{45}$) carried out for a unidirectional layer of the viscoelastic HSM, compared to a reference simulation with the heterogeneous microstructure show the accuracy of the approach and the anisotropy of the effective material model for the roving (see Fig. 7).

![Fig. 10. Validation of the viscoelastic homogenisation: a) Meshed microscopic RVE; b) Verification setup; c) Comparison reaction forces](image)

For the numerical simulation of the behaviour of the whole composite according to Fig. 11, a geometric model of the mesoscopic textile reinforcement is needed. Because the inner structure of the consolidated textile fabric is very complex, it is not possible to directly use information from CT scans or micrograph images.

Therefore, an idealized geometric model of the mesoscopic RVE has been generated (see Fig. 11 b). Based on representative 2D sections from the CT scan (see Fig. 11 a), the dimensions of the roving (black and white lines) and the height of the layers have been determined (0.5 mm). For the simulation of the composite, several of these RVE were assembled to form a quarter model of the specimen with the symmetry planes between vector 1&2 and 2&3 (see Fig. 11 c).

![Fig. 11. Development of the geometric model; a) CT scan; b) RVE; c) 1/4-part of the geometric model of the composite specimen](image)

**Simulation of the long-time behaviour**

The simulations of a creep and a relaxation load case considering compressive loading applied to the composite (CC|C|117MPa, CC|R|4%) are in good agreement with the corresponding experimental results.

![Fig. 12. Comparison of experimental results for static compression tests](image)
While the creep simulation predicts a slightly softer effective material behaviour than indicated by the experiments, the simulated relaxation behaviour appears to be stiffer than the real composite specimen. Possible reasons for that could be deviations between the geometry of the finite element model and the real textile reinforcement structure of the specimen, the assumed fibre volume fraction of the roving in the homogenisation procedure as well as the tension/compression asymmetry of the matrix material.

Based on the experiments, a material model for PP was chosen and its parameters were identified from the experimental data. In order to predict the effective material behaviour of the composite, a two-step homogenisation was performed. In the first step (micro-meso) the effective anisotropic, viscoelastic behaviour of areas with high fibre volume fraction, the so-called roving, has been computed using homogenisation procedures adapted to linear viscoelastic material behaviour. For the second step (meso-macro) an idealized geometric model of the local textile reinforcing structure was deduced from CT scans. Using this mesoscopic RVE model, the behaviour of the composite was simulated and compared to static and long-term compression experiments carried out with composite specimens.

Although the overall characteristics of the experiments are represented by the numerical results, the exact quantities of the experiments cannot be matched. Possible reasons for that could be deviations between the geometric model and the real structure of the composite or the recognized asymmetry between tension and compression of the matrix material.

Future work therefore has to focus on a better description of the tension/compression asymmetry in the behaviour of the matrix material and an automated procedure to obtain a geometric RVE model directly from a CT scan.

In addition, for life time analysis of the material, long-term experimental data up to $10^8$ s are necessary. Standard testing machines, used in the research described here are not suitable for that. Therefore, a long term test rack is currently under development.

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