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Hygrothermal viscoelastic material characterisation of unidirectional continuous carbon-fibre reinforced polyamide 6

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Abstract

This paper presents results of material characterisation experiments on the hygrothermal viscoelastic behaviour of unidirectional laminates of continuous carbon-fibre reinforced polyamide 6. The material behaviour when subjected to the automotive painting process is of interest. Coefficients of thermal- and -moisture expansion were determined from dilatometer experiments and micrometer measurements together with weighing, respectively. Diffusion coefficients were generated from thermogravimetric analysis and fitted with the Arrhenius equation. Dynamic mechanical analysis and digital image correlation of quasi-static tensile tests were performed to obtain a relaxation curve and a major Poisson's ratio, respectively. The Williams-Landel-Ferry equation was fitted to the time shift factors.

Keywords: A. Carbon fibre, A. Thermoplastic resin, B. Thermomechanical, B. Rheological properties, X. Hygroscopy

1. Introduction

The application of composite materials in the body structure of passenger vehicles has seen a gradual increase over the past few years due to the efforts made in the automotive industry to restrain the trend of increasing kerb weight [1, 2, 3]. Structural composite parts in the automotive industry most often feature a thermoset matrix due to lower raw materials costs in comparison to a thermoplastic equivalent [4]. However, legislative changes to the requirements

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on recyclability of newly produced vehicles have led to the interest in utilising polymers with a thermoplastic nature because of the possibility to recycle [5]. Contrarily, composites made of thermoset matrix material cannot be remelted because of cross-linking [6, 7]. The matrix- and fibre material of a continuous fibre-reinforced thermoplastic can be separated by means of melting since a melting temperature exists for thermoplastics. Nevertheless, recyclable composites are often still landfilled due to the difficulties in physically recovering them due to strong connections to other parts [8].

In addition to providing recyclability advantages, thermoplastic composites benefit from rapid processing, improved fracture toughness and joining possibilities through welding [9, 10, 11, 12, 13]. It must be kept in mind that adapting the conventional automotive process chain of existing vehicle models to optimise the manufacturing process for thermoplastic matrix materials is not sensible from an economical perspective. It must therefore be noted that composite parts are connected to the body-in-white (BIW) in the body shop, which occurs prior to entering the paint shop. The consequence is that structural parts with a thermoplastic matrix, intended for existing vehicle models, are subjected to the automotive paint shop. The BIW is passed through a series of drying ovens during the automotive paint shop in which a maximum temperature up to approximately 190 $°C$ is encountered [14, 15]. This temperature surpasses the glass transition temperature of economically sensible thermoplastic matrix materials and the deformation behaviour must be investigated to be able to safeguard the intended geometry and functioning of these structural parts.

The deformation of polymer composites is governed by thermal- and hygroscopic expansion in combination with clamped boundary conditions [16, 17]. The automotive painting process is of special interest due to the deformation that is caused by the imposed thermal loads. The material characterisation techniques in this study were carried out with polyamide 6 (PA6) as matrix material and were aimed at identifying all necessary material parameters to accurately model the deformation behaviour of thermoplastic composite structural parts during the automotive painting process. It is noted that PA6 is a hydrophilic polymer and absorbs moisture by diffusion [18, 19, 20]. Hence, composite structural parts with PA6 as matrix material display an increase in moisture concentration as a result of the pre-treatment and cathodic dip painting process. It follows that these parts enter the dryers with a non-zero

moisture content. The moisture diffusion of various polyamides is often conform to Fickian behaviour and can be modelled accordingly [21, 22, 23].

Additionally, thermoplastic materials show some polymer-specific phenomena. Recrystallisation occurs during the application of the thermal load and results in a higher degree of crystallinity that increases the density of the polymer and yields post-shrinkage [24, 25, 26]. Specifically to PA6, the application of a thermal load also causes a transition of the polymer crystals with a γ-form to an α-form, leading to an increase in density as well [27, 28]. The effects of recrystallisation and change in crystal form can be modelled by utilising temperature-dependent coefficients of thermal expansion (CTE), analogous to the approach taken by Heinle and Drummer [29]. Baran et al. also report the importance of including shrinkage due to crystallisation in the modelling of composite manufacturing processes [30]. The determination of a CTE is often carried out by means of a dilatometer, as demonstrated by Gabrion et al. [31]. Characterising the viscoelastic properties of a thermoplastic composite is often performed through dynamic mechanical analysis (DMA) [31, 32, 33, 34, 35, 36]. Ma et al. proposed a method to measure hygroscopic expansion by means of DMA [37]. Mechanical models to describe the long-term stress relaxation behaviour of structural members are presented by Ascione et al. [38] and Berardi and Mancusi [39, 40]. The long-term creep behaviour of glass-fibre-reinforced polymers (GFRP) was studied by Berardi et al. [41] and successfully modelled by means of Burger's model. It was concluded that long-term creep effects are relevant in GFRP laminates manufactured with an epoxy matrix. Long-term stress relaxation behaviour of thermorheologically simple materials can also be described by experimental data obtained during short-term testing through the time-temperature superposition principle [42].

This paper presents the results of several material characterisation methods that have been carried out with specimens taken from unidirectional (UD) laminates of continuous carbon-fibre reinforced PA6. The objective of the study is to identify the hygrothermal- and mechanical response of these laminates. This information will be used later as input for a numerical model to simulate the deformation behaviour of multi-directional laminates, manufactured with identical material, subjected to the automotive painting process.

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2. Material and methods

Various experiments have been carried out to identify the hygrothermal viscoelastic material behaviour of the UD carbon-fibre reinforced PA6 for which the details are provided (See Section 2.1). All investigated specimens were conditioned to either a fully dry state or to a saturated moisture content, depending on the requirement as per material characterisation technique. The conditioning was carried out according to the procedure that is described in DIN EN ISO 1110. Drying the specimens was performed in an oven at 80 $^\circ C$, whereas conditioning the specimens to a state of moisture saturation occurred in an environmental chamber at 70 $^\circ C$ and 62% relative humidity. The glass transition temperature of the investigated PA6 in completely dry state was measured by means of differential scanning calorimetry on a DSC 204 F1 Phoenix$^\circledR$ and was found to be 67 $^\circ C$. Cutting the specimens to the correct dimensions, suitable for carrying out the material characterisation, was performed by waterjet cutting to prevent thermally loading the specimens and altering the degree of crystallinity prior to testing.

2.1. UD carbon-fibre reinforced PA6

Specimens were taken from UD laminates that had been produced with Celstran$^\circledR$ CFR-TP PA6-CF60-01 from Celanese Chemical Europe GmbH; a carbon fibre-reinforced UD tape with PA6 as matrix. The manufacturing of the laminates was carried out at the Fraunhofer IGCV in Augsburg, Germany. The UD tapes had been stacked unidirectionally with a FiberForge$^\circledR$ Relay 2000 and were consolidated by means of hot compression moulding after infrared pre-heating. The pre-heating was performed to reduce the total time required for consolidation. A total of 14 layers of UD tape had been used to create the UD laminates and resulted in a nominal thickness of 1.7 mm. The fibre volume content of the produced UD laminates was analysed by matrix burn-off in combination with thermogravimetric analysis (TGA) and yielded a value of 47.8% $\pm$0.2%. The short-term modulus of the UD laminates were tested in both longitudinal- and transverse directions according to DIN EN ISO 527-4 as preliminary research. The results are shown in Table 1.

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2.2. Dilatometer experiments

The thermal expansion of the UD laminate was investigated on a Netzsch DIL 402 C dilatometer. This dilatometer has a strain measurement resolution of 0.625 nm and the specimens were cut to a size of 100 x 8 x 1.7 mm^3. All specimens concerning the dilatometer experiments had been conditioned to a dry state to eliminate the effect of hygroscopic shrinkage when subjecting the specimens to the temperature profile of interest. A heating- and cooling rate of 2 $°C/min$ were chosen for a temperature range from 25 $°C$ to 200 $°C$. The results of the measurements are strain- and temperature as a function of time. Eq. (1) is used to determine the coefficients of thermal expansion [43].

$$
\begin{aligned}
\alpha_{11} &= \frac{\partial \varepsilon_{11}}{\partial T} \\
\alpha_{22} &= \frac{\partial \varepsilon_{22}}{\partial T}
\end{aligned}
\tag{1}
$$

It is noted that these are tangential coefficients. UD specimens have been measured for elongation in both 0°- and 90° directions. The applied temperature profile and the measured strains are shown in Fig. 1 and Fig. 2 for the 0°- and 90° fibre directions, respectively.

2.3. Micrometer measurements and weighings

The hygroscopic expansion of the UD laminate was analysed in both 0°- and 90° directions by carrying out micrometer measurements and weighings on an analytical scale. The utilised micrometer and analytical scale were the Atorn Keeptronic with a resolution of 0.01 mm and a VWR LA Series with a resolution of 0.1 mg, respectively. Three specimens for both measurement directions with dimensions of 100 x 8 x 1.7 mm^3 were first conditioned to a dry state before placing them in the environmental chamber to absorb moisture. Initial weight- and length measurements were carried out to obtain reference values for the completely dry UD laminate. Two measurements were taken on the first day after which the interval was changed to once per day until moisture saturation was observed. Eq. (2) is applied to calculate the coefficients of moisture expansion (CMEs).

$$\beta_{11} = \frac{\partial \varepsilon_{11}}{\partial M}$$
$$\beta_{22} = \frac{\partial \varepsilon_{22}}{\partial M} \tag{2}$$

The moisture content that is indicated with M in Eq. (2) was determined with the weight measurements of the specimens as shown in Eq. (3). In Eq. (3), $W(t)$ and W_0 denote the mass at moment t in time and the initial dry weight, respectively.

$$M(t) = \frac{W(t) - W_0}{W(t)} \tag{3}$$

2.4. Thermogravimetric analysis

The rate of moisture diffusion was assessed by performing thermogravimetric analysis on a Netzsch TG 209 F1 Libra and determining diffusion coefficients afterwards. The specimens that were investigated by means of TGA were first conditioned to a state of moisture saturation. The decreasing weight of the specimens was then measured as they were subjected to an ambient air with temperature values of 80 °C, 120 °C, 150 °C, and 200 °C. The coefficients of diffusion have been determined through the application of an equation that is valid for one-dimensional mass diffusion. Hence, the cutting edges of the specimens were sealed to minimise moisture egress through these sides. The weight loss for each specimen can be computed with Eq. (4) and is required for the determination of the diffusion coefficients.

$$G(t) = \frac{W(t) - W_0}{W_\infty - W_0} \tag{4}$$

The symbols W_0 and W_∞ in Eq. (4) represent the mass of the specimen at the start and end of the TGA measurement, respectively. The value of W_∞ is identical to the completely dry mass of the specimen. The weight loss can be plotted as a function of time and was fitted with the expression shown in Eq. (5), again noting that this expression is only valid for one-dimensional diffusion through an infinitely large plate of which the concentration at the surfaces are kept constant [44, 22]. The only fitting parameter in this expression is the

diffusion coefficient, denoted with D. The distance of diffusion is governed by l in Eq. (5) and was taken to be equal to the half-thickness of the specimen due to moisture egress out of both major surfaces of the specimens.

$$G(t) \approx 1 - \frac{8}{\pi^2} \sum_{k=0}^{N} \frac{1}{(2k+1)^2} \cdot e^{-(2k+1)^2 \pi^2 D \cdot t \cdot l^{-2}} \tag{5}$$

Fitting the weight loss data to Eq. (5) yields diffusion coefficients that correspond to the particular temperature of the corresponding TGA measurement. The Arrhenius equation, displayed in Eq. (6), was used to fit the discrete set of diffusion coefficients to obtain a continuous function of the diffusion coefficient for the entire temperature range [45]. The symbols D_0 and E_A in Eq. (6) represent the frequency factor and activation energy, respectively.

$$D(T) = D_0 \cdot e^{\dfrac{E_A}{R \cdot T}} \tag{6}$$

2.5. Dynamic mechanical analysis

The stress relaxation behaviour of the UD laminate was analysed by means of DMA on a Netzsch EPLEXOR 500 N. Specimens were trimmed to a size of 80 x 10 x 1.7 mm^3 to facilitate a tensile measuring mode. Frequency sweep measurements were performed for a temperature range of 20 $°C$ to 220 $°C$ with a step size of 10 $°C$. The measured frequencies range from 0.3 Hz to 30 Hz with 8 steps per decade. A trade-off was made here between the duration of the measurement and not damaging the specimen. A hold period of 20 minutes was included between consecutive frequency sweeps to ensure thermal stability inside the DMA test chamber. The stress relaxation was expected to be most significant in 90° direction and DMA analysis was thus only conducted in this direction.

The objective of the DMA was to obtain the transverse relaxation modulus at one specific reference temperature. A mastercurve was constructed for the storage modulus by shifting the individual measurements of the storage modulus on the logarithmic time scale to form one continuous curve, applying the principle of TTS [42]. A reference temperature of 20 $°C$ was chosen for the construction of the mastercurve. The shift factors required for the

construction of the mastercurve also provide quantitative information on how temperature influences the rate of stress relaxation. The objective simulation covers a large temperature range and thus the correct modelling of the temperature-dependent rate of stress relaxation is important to predict accurate deformations. The mastercurve is shown in Fig. 3 and was fitted to a generalised Maxwell model which was formulated in the frequency domain as shown in Eq. (7) [46].

$$E'(\omega) = E'_\infty + \sum_{i=1}^{N} \frac{E'_i \omega^2 + \tau_i^2}{1 + \omega^2 \tau_i^2} \tag{7}$$

The symbols E'_∞, E'_i, and τ_i denote the long-term modulus and the modulus and relaxation time of the ith Maxwell element. The values of the relaxation time of all Maxwell elements were kept constant and evenly spaced on the logarithmic scale. Fitting the moduli of the Maxwell elements was performed with the genetic algorithm method that is provided in the Global Optimization Toolbox in MATLAB. Utilising 32 Maxwell elements led to a root-mean-square (RMS) error of 26.7 MPa. The RMS error has a magnitude of 4.9% of the smallest measured storage modulus and the fit shown in Fig. 4 is thus considered to be satisfactory. The values of τ_i and the corresponding values of E'_i can be used in the generalised Maxwell element formulated in the time domain, Eq. (8). The obtained curve from Eq. (8) represents the relaxation modulus of the UD laminate in 90° direction.

$$E_{22}(t) = E'_\infty + \sum_{i=1}^{N} E'_i e^{-t/\tau_i} \tag{8}$$

The time shift factors that were obtained by constructing the mastercurve of the storage modulus are utilised to fit the Williams-Landel-Ferry (WLF) equation and analyse the temperature-dependency of the stress relaxation behaviour. The WLF equation is shown in Eq. (9) in which the terms C_1 and C_2 denote the fitting parameters. The symbols a_T and T_0 denote the time shift factors and the reference temperature, respectively.

$$\log a_T(T) = -\frac{C_1\,(T - T_0)}{C_2 + T - T_0} \tag{9}$$

2.6. Digital image correlation

Quasi-static tensile tests were performed to obtain the major Poisson's ratio of UD specimens at room temperature. The specimens and testing conditions were chosen to be in agreement with DIN EN ISO 527-4. The dimensions of the specimen measured 250 x 15 x 1.7 mm^3 and the crosshead speed was set to 2 $mm \cdot \text{min}^{-1}$. End tabs of cross-ply GFRP with an orientation of $\pm 45°$ were applied to increase gradual load introduction and reduce the chances of damaging the specimen in the clamping area. A Zwick/Roell Z100 universal testing machine was used in combination with digital image correlation (DIC) to measure the longitudinal- and transverse strains. The DIC setup included the ARAMIS system with the Deformation 2.3M sensor that has a resolution of 1936 x 1216. The utilised lens allowed a measuring volume of 140 x 95 x 90 mm^3 and was calibrated at a temperature of 23.0 $°C$ with a camera angle of 25.492°. A picture of the experimental setup for the DIC measurement is shown in Fig. 5. The major Poisson's ratio was evaluated for six specimens at longitudinal strain values of 0.4% and 0.5% with Eq. (10).

$$\nu_{12} = -\frac{\varepsilon_{22}}{\varepsilon_{11}} \tag{10}$$

3. Results and discussion

3.1. Coefficients of thermal expansion

The CTEs that have been calculated with Eq. (1) are shown in Fig. 6 and Fig. 7 for 0°- and 90° fibre orientations, respectively. The variation between the CTEs for heating and cooling originates from two phenomena that are present because of the application of the thermal loading. It is assumed that the transition of the PA6 crystals from γ-form to α-form is one of the reasons for the increase in density of the laminate. This assumption is based on the relatively low cooling rates in the dilatometer tests which favours the presence of polymer crystals with an α-form as opposed to a γ-form [28]. Secondly, recrystallisation is activated

as the specimens are heated above the glass transition temperature. Both phenomena would explain the decline in strain during both temperature hold periods in the curve that is depicted in Fig. 2 for 90° direction. Furthermore, these phenomena result in a net shrinkage that can be observed by the negative strain values shown at the end of the cooling phases in Fig. 2. Both phenomena produce differences in CTEs for the heating- and cooling phases, as shown in Fig. 7. It is remarkable that the strain of the UD laminate in 0° direction, shown in Fig. 1, does not return to zero after cooling. This behaviour is assumed to be caused by the fibres as the strain in this direction is highly fibre-dominant due to the high stiffness of the fibres in comparison to the matrix. The importance of determining separate CTEs for heating- and cooling phases for accurate simulation purposes is again noted.

3.2. Coefficients of moisture expansion

The measured hygroscopic strain is plotted versus the moisture concentration of the specimens that have been weighed for the determination of the CMEs in Fig. 8. The results of the 0°- and 90° fibre directions are shown on the left- and right hand sides, respectively, with corresponding CMEs of $5.58e^{-3}$ and $4.26e^{-1}$. These are determined by means of a linear fit through the origin. It is noted that the degree of experimental scatter is large for the measurements in 0° direction. The scatter is caused by measurement error that is introduced by the resolution of the micrometer in combination with the relatively small hygroscopic strains in 0° direction.

3.3. Diffusion coefficients

Fig. 9 displays the fitted Arrhenius function, of which the expression is shown in Eq. (6), to the determined diffusion coefficients (See Section 2.4). It is noted that the high goodness of fit indicates that the Arrhenius equation provides an excellent method of describing the temperature dependency of the one-dimensional diffusion coefficient.

3.4. Stress relaxation behaviour

The relaxation curve, that is constructed with the moduli of the Maxwell elements in Eq. (7), is displayed in Fig. 10. It is noted that the relaxation curve is valid only for the reference temperature of 20 °C. The resulting instantaneous modulus of the relaxation curve

is in proper agreement with the values measured by quasi-static tensile tests carried out at room temperature. The instantaneous modulus that was obtained from the quasi-static tensile tests was calculated with the stress values measured at 0.05%- and 0.25% strain, in agreement with DIN EN ISO 527-4. The specimen relaxes during the strain application, which takes approximately 15 seconds, and this explains the discrepancy between the measured instantaneous modulus with quasi-static tensile tests and the predicted instantaneous modulus by means of DMA. The temperature dependency of the transverse relaxation modulus can be described by means of the WLF equation, given in Eq. (9). The coefficients C_1 and C_2 in Eq. (9) have been determined by fitting the WLF equation to the time shift factors obtained from applying the TTS to construct the mastercurve of the storage modulus. This fit is shown in Fig. 11 and the values for C_1 and C_2 are 69.85 and 254 $°C$, respectively.

3.5. Major Poisson's ratio

The values of the major Poisson's ratio that had been determined for six specimens at strain values of 0.4% and 0.5% are shown in Table 2. The average major Poisson's ratio of all measurements is equal to 0.400 and the standard deviation is found to be 0.024. It is observed that the major Poisson's ratio decreases at higher strain magnitudes due to relaxation of the matrix constituent.

4. Conclusion

The hygrothermal viscoelastic material behaviour of UD laminates of continuous carbon-fibre reinforced PA6 was analysed by means of various experiments. Thermal- and hygroscopic expansion behaviour was measured by performing dilatometer experiments and micrometer measurements in combination with weighing, respectively. The thermal- and hygroscopic expansion was quantified by the determination of coefficients of thermal- and -hygroscopic expansion, respectively. A distinct post-shrinkage was observed for the UD laminate with a fibre orientation of 90° and also the UD laminate with a fibre orientation of 0° did not return to zero strain after removal of thermal loading. The rate of moisture diffusion out of the moisture saturated specimens that were dried at various constant temperature conditions was investigated by TGA. The Arrhenius equation was used to describe the temperature dependency of the experimentally obtained diffusion coefficients and a proper fit to the

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experimental data was obtained. The stress relaxation behaviour in transverse direction of the UD laminate was measured by multiple frequency sweeps under isothermal conditions with DMA. Fitting the constructed mastercurve by a generalised Maxwell model with 32 terms yielded a RMS error of 4.9% of the smallest measured storage modulus. The WLF equation was utilised to describe the temperature-dependency of the relaxation behaviour. Finally, DIC was applied to quasi-static tensile tests to identify the major Poisson's ratio of the UD laminate at room temperature to be 0.400.

It was concluded that the post-shrinkage and temperature-dependent relaxation behaviour of the studied material impose challenges for use in structural automotive applications and therefore demand further investigation to validate the accuracy of simulation approaches. The present material characterisation will hence be used for numerical analyses of the influence of the automotive painting process on the deformation behaviour of multi-directional laminates.

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Fig. 1. Applied temperature profile for dilatometer experiment and 0° strain measurement.

Fig. 2. Applied temperature profile for dilatometer experiment and 90° strain measurement.

Fig. 3. Constructed mastercurve of storage modulus obtained from DMA measurement in 90° direction.

Fig. 4. Fitted generalised Maxwell model to the mastercurve shown in Fig. 3.

Fig. 5. Experimental setup for measuring the major Poisson's ratio by means of DIC.

Fig. 6. Calculated tangential CTEs of the dry composite material in 0° direction for two heat cycles.

Fig. 7. Calculated tangential CTEs of the dry composite material in 90° direction for two heat cycles.

Fig. 8. Strain and moisture concentration results of the micrometer measurements and weighings, including the determined coefficients of moisture expansion.

Fig. 9. Fitted Arrhenius curve that describes the diffusion coefficient as a function of temperature.

Fig. 10. Resulting relaxation curve from the fitting parameters of the generalised Maxwell model to fit the mastercurve shown in Fig. 4.

Fig. 11. Fitted WLF equation to describe the time shift factors as a function of temperature.

Table 1. Experimentally determined short-term properties of Celstran® CFR-TP PA6-CF60-01 at room temperature.

Specimen	E_{11}	Specimen	E_{22}
1	100.05 GPa	6	4.23 GPa
2	107.20 GPa	7	4.43 GPa
3	109.75 GPa	8	4.14 GPa
4	110.32 GPa	9	4.45 GPa
5	102.34 GPa	10	4.21 GPa

Table 2. Major Poisson's ratio values of Celstran® CFR-TP PA6-CF60-01 measured with DIC at room temperature.

Specimen	ν_{12} at 4% strain	ν_{12} at 5% strain
1	0.382	0.370
2	0.427	0.416
3	0.420	0.418
4	0.392	0.369
5	0.427	0.422
6	0.396	0.362