Rheological characterisation and modelling of a glass mat reinforced thermoplastic for the simulation of compression moulding

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Abstract. The use of hybrid components in the automotive industry is steadily increasing due to their lightweight potential. By combining fibre-reinforced plastics with metallic materials, high-strength components with lower weight than monolithic metal parts can be realised. The overall aim of the project "HyFiVe" is to exploit this potential for electric vehicles by developing a scaled battery housing structure made of a glass mat reinforced thermoplastic (GMT) paired with unidirectional reinforced (UD) tapes and a metallic reinforcement frame. The GMT is formed by compression moulding and serves as the base of the battery housing structure. Numerical simulation is an efficient tool for process design that can determine a suitable process window and reduce experimental trial-and-error tests. Particularly, realistic modelling of the GMT flow behaviour is essential for reliable simulation results. In this contribution, the rheological properties of a GMT consisting of a polyamide 6 (PA6) matrix with 30% glass fibre reinforcement were determined. Isothermal compression tests were carried out with a parallel plate rheometer at different temperatures and varying squeeze rates. The squeeze force and punch displacement were evaluated to determine the rheological data of the GMT. Two methods for the modeling of the flow behaviour were considered. At first, pure shear flow was assumed and the viscosity was modelled as a function of the shear rate by means of a power-law. Secondly, a pure biaxial extension was assumed and the true stress was modelled in dependence of true strain and strain rate. Subsequently, for a verification of the material models, the compression tests were simulated in ABAQUS using the Coupled Eulerian-Lagrange (CEL) approach and the results were compared to the experiments.

Introduction

Due to the high demand for lightweight construction, conventional materials are increasingly substituted by materials with higher strength. The utilisation of fibre-reinforced plastics (FRP) plays a decisive role in this context [1]. FRP with a thermoplastic matrix are available as pre-impregnated semi-finished products in the form of long-fibre-reinforced thermoplastics (LFT) or glass mat-reinforced thermoplastics (GMT). The latter consists of a fibre mat with randomly arranged continuous fibres, which are impregnated with the matrix material. GMT is characterised by a high specific strength and specific energy absorption capabilities, which makes it particularly suitable for lightweight components in the automotive sector [2]. Application examples include bumper beams, spare wheel wells and battery trays. Further advantages are recyclability and noise cancellation [3]. GMT is also characterised by comparatively simple processing, as forming is usually carried out by compression moulding using standard hydraulic presses. In addition to simple forming, GMT can be used in combined processes. Behrens et al. developed a thermoforming process for the combined forming of organic sheet and GMT [4]. The GMT was used to integrate complex geometric elements into the component. As the forming could be carried out in one stroke, no additional process steps were necessary. The development of a combined forming process is also the objective of the project "HyFiVe", in which a battery housing structure is formed from GMT, UD tapes and a metallic reinforcement frame. The GMT serves as the basic structure holding the battery modules and cables [5]. The application of numerical simulation methods is essential for efficient process design, as it allows a process window to be identified that ensures the production of good parts. As a result, trial and error testing can be reduced, saving costs, time and materials. However, numerical process design requires detailed knowledge of the material behaviour to obtain realistic simulation results. Especially the rheological data of GMT has to be characterised and modelled to simulate the flow behaviour in the mould cavity.

In contrast to short-fibre-reinforced plastics, the rheological data of GMT cannot be measured in a capillary rheometer, as this would require the semi-finished product to be trimmed, which would shorten the fibres. For this reason, the use of a parallel plate rheometer has been established in literature [6]. With this procedure, a circular specimen of the material is compressed between two parallel plates at a constant squeeze rate and temperature up to a defined final thickness, with the polymer melt flowing in radial direction. During the test, the applied squeeze force and displacement are recorded in order to determine the rheological properties of the material. The test is characterised by its simplicity and is very similar to the compression moulding process, which explains why it is widely used in literature to characterise thermoplastic melts with long fibres. The incompressibility of the material and therefore a constant volume is usually assumed for the evaluation. When modelling the flow behaviour, a distinction is made between two extreme cases. In the first case, which is shown in Fig. 1 a), pure shear flow is assumed. In this case, pure sticking of the material to the tool surface is assumed. In the second flow form, which is shown in Fig. 1 b), a pure biaxial extension of the material is assumed, which is often referred to as a “plug flow”. In this case, the material slips completely off the tool surface.

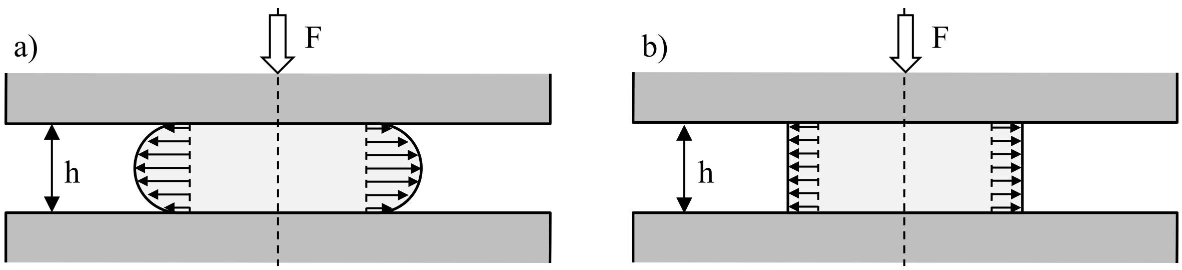

Fig. 1. Pure shear flow a) and pure biaxial extension b) in a parallel plate rheometer.

The first mathematical modelling approach of the material flow in the parallel plate rheometer were published by Scott, who investigated the physical relationships [7]. Under the assumption of a pure shear flow (Fig. 1 a), the squeeze force F_{Shear}, which is necessary to move the plate at a constant velocity u, can be determined using the Scott equation.

$$F_{Shear}=\left(\frac{u}{4}\right)^n\left(\frac{2n+1}{n}\right)^n\left(\frac{2}{h}\right)^2\frac{K\pi R^{n+3}}{(n+3)} \quad (1)$$

Here, h stands for the plate distance or the current height of the specimen and R for the radius of the specimen. K and n are the coefficients of a power law that defines the shear viscosity η_{Shear} as a function of the shear rate $\dot{\gamma}$.

$$\eta_{Shear}=K\dot{\gamma}^{n-1} \quad (2)$$

Under the assumption of a pure biaxial extension of the material (Fig. 1 b), the required squeeze force F_{ext} can be determined according to Kotsikos using Eq. 3 [8].

$$F_{ext}=\pi R^2 A \left(\frac{u}{h}\right)^m \tag{3}$$

Accordingly, the coefficients A and m can be used to calculate the extensional viscosity η_{ext} of the material as a function of the strain rate $\dot{\varepsilon}$.

$$\eta_{ext}=A\dot{\varepsilon}^{m-1} \tag{4}$$

However, Kotsikos found that the coefficient A is not constant, but depends on the strain [8]. To take the strain into account, the biaxial extension can also be modelled using the true stress σ as a function of the true strain ε and strain rate $\dot{\varepsilon}$ [9].

$$\sigma=\frac{F_{ext}}{A}=\frac{F_{ext}}{\frac{A_0 h_0}{h}}=\frac{F_{ext}h}{\pi R^2 h_0}, \qquad \varepsilon=\ln\left(\frac{h}{h_0}\right), \qquad \dot{\varepsilon}=\frac{u}{h} \tag{5}$$

Using this method, the extensional viscosity can be calculated according to the relationship in Eq. 6 [9].

$$\eta_{ext}=\frac{\sigma}{\dot{\varepsilon}} \tag{6}$$

Kotsikos et al. carried out isothermal compression tests in a parallel plate rheometer with circular specimen made of GMT with polypropylene (PP) as matrix material [8]. The material flow was modelled both as pure shear flow and pure biaxial extension. As a result, the assumption of a pure shear flow led to inadequate results regarding the predicted squeeze forces, which were significantly underestimated compared to the experimental results. In contrast, the assumption of a pure extensional flow provided better agreement with experimental results. Dweib and Ó Brádaigh also carried out compression tests with GMT made of PP [10]. The originally circular specimen showed an elliptical shape after forming. Due to the anisotropy, two viscosity models were modelled depending on the main flow directions under the assumption of a pure extensional flow. Scanning electron microscopy images showed that more fibres were arranged along one main fibre direction during forming. A significant anisotropy of the viscosity of GMT was also found by Dörr et al., which was attributed to a predominantly in-plane fibre orientation [11]. By taking the anisotropy in the material modelling into account, a higher prediction accuracy could be achieved in the numerical simulation, as it was possible to predict wrinkling during forming. Thattaiparthasarthy et al. carried out isothermal compression tests with a long-fibre-reinforced thermoplastic made of PP and investigated the effects of temperature, fibre length and fibre weight fraction on the viscosities [6]. An increase in temperature and a decrease in fibre length and fibre weight fraction led to a reduction of the viscosity. Huang et al. used a parallel plate rheometer to analyse GMT with PA6 as matrix material [12]. For the modelling, a pure shear flow was assumed and the viscosity was modelled as a function of the shear rate. The material data was used to simulate the compression tests in Moldex3D. However, the resulting squeeze force was significantly underestimated in the simulation. The separation of fibre and matrix was stated as the reason for the deviations. Behrens et al. also characterised GMT with PA6 as matrix material. They assumed a biaxial extension of the material and modelled the flow by means of true stress as a function of true strain and strain rate [13]. In a numerical simulation, good agreement was achieved regarding the predicted material flow and the pressure distribution compared to the experiments. The objective of this contribution is the characterisation of a GMT with a PA6 matrix using a parallel plate rheometer to model the material flow in the simulation of compression moulding. Since different assumptions and thus modelling approaches can be found in literature [4-8], both a pure shear flow and a pure extensional flow are assumed for the evaluation of the rheological data. Subsequently, the modelling methods are used in a numerical simulation of the compression tests to identify the approach with the highest prediction accuracy. Due to the high deformations that occur during compression moulding, the CEL approach is used for the simulation.

Experimental Test Setup

For the compression tests, a parallel plate rheometer made of C45 with a diameter of 200 mm was used (Fig. 2). The characterised material was a GMT consisting of a PA6 matrix and a glass fibre reinforcement with a volume fraction of 30%. The glass fibres had an average length of 50 mm and where randomly oriented. The material was provided by Mitsubishi Chemical Advanced Materials AG in the form of sheets with a thickness of 4.4 mm. Circular specimens with a diameter of 50 mm were prepared by water jet cutting. The tests were carried out isothermally at process-relevant temperatures of 240 °C, 260 °C and 280 °C. Therefore, the plates were heated to the corresponding temperatures by means of integrated heating coils.

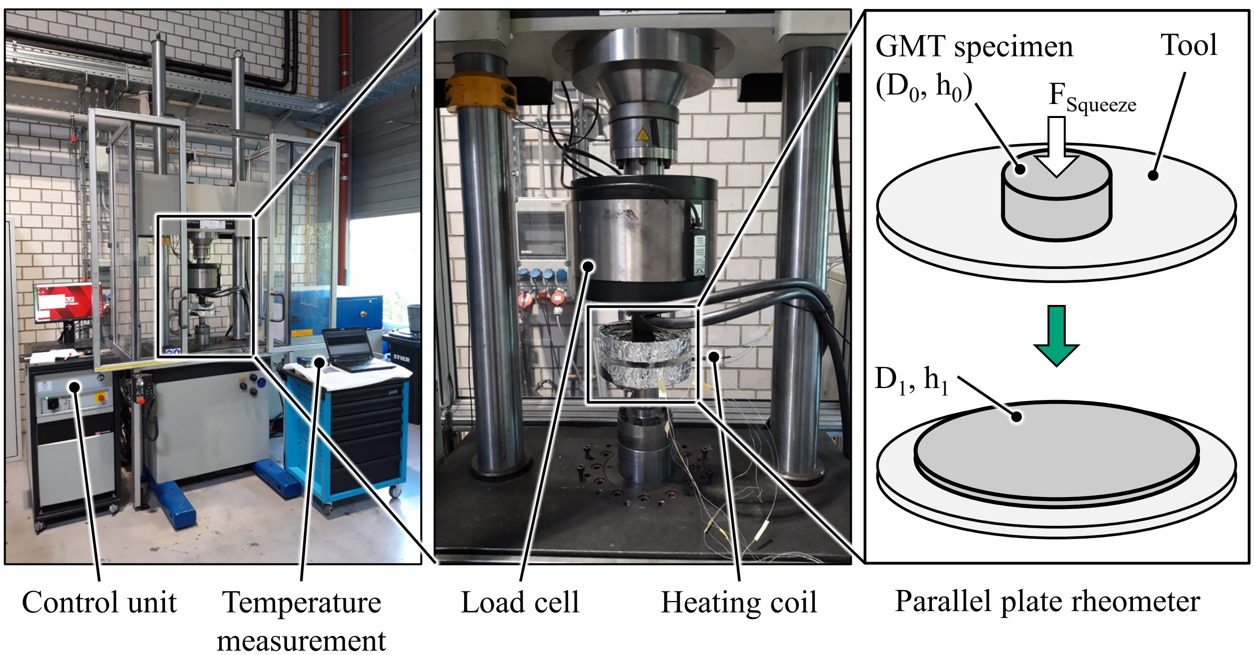

Fig. 2. Experimental setup for the compression tests.

Three circular specimens were placed on top of each other between the tool plates, which were then brought into contact. The number of specimens and thus the total height of 13.2 mm was chosen based on the planned layer setup in the later compression moulding tests on industrial scale. Analogous to [12], an aluminium foil was inserted between the samples and the parallel plates, to prevent the GMT from sticking to the tool surfaces. After insertion, the specimens were heated to the corresponding test temperature by conduction through the plates. The required heating time was determined beforehand using thermocouples, which were inserted between the GMT specimen during heating. After the required holding time was reached, the plates were moved together at a constant squeeze rate to a minimum tool gap of 2.7 mm, whereby the GMT flowed in radial direction. Four squeeze rates were applied (0.02 mm/s, 1 mm/s, 5 mm/s, 25 mm/s) and each test was performed three times. During the tests, the squeeze force and punch position were recorded to determine the rheological properties of the GMT in the following sections.

Experimental Results

Fig. 3 shows the measured squeeze forces over the punch displacement for the investigated squeeze rates and temperatures. At the beginning of the compression process, only very low squeeze forces occurred. Subsequently, after a displacement of approx. 2 mm, the squeeze force increases almost linearly during the compression test. From a stroke of approx. 8 mm, the gradient increases exponentially due to the compaction of the fibre bed until it reaches its maximum at a displacement of 10.5 mm [14]. It can be seen that the required forming force increases with higher squeeze rates. Furthermore, higher forming temperatures lead to a decrease of the measured squeeze forces. The geometries of the specimen are shown in Fig. 3 b) before and after forming for two experimental tests. It can be seen, that the shape after forming remains approx. circular, so that an isotropic flow behaviour is assumed in the material modelling.

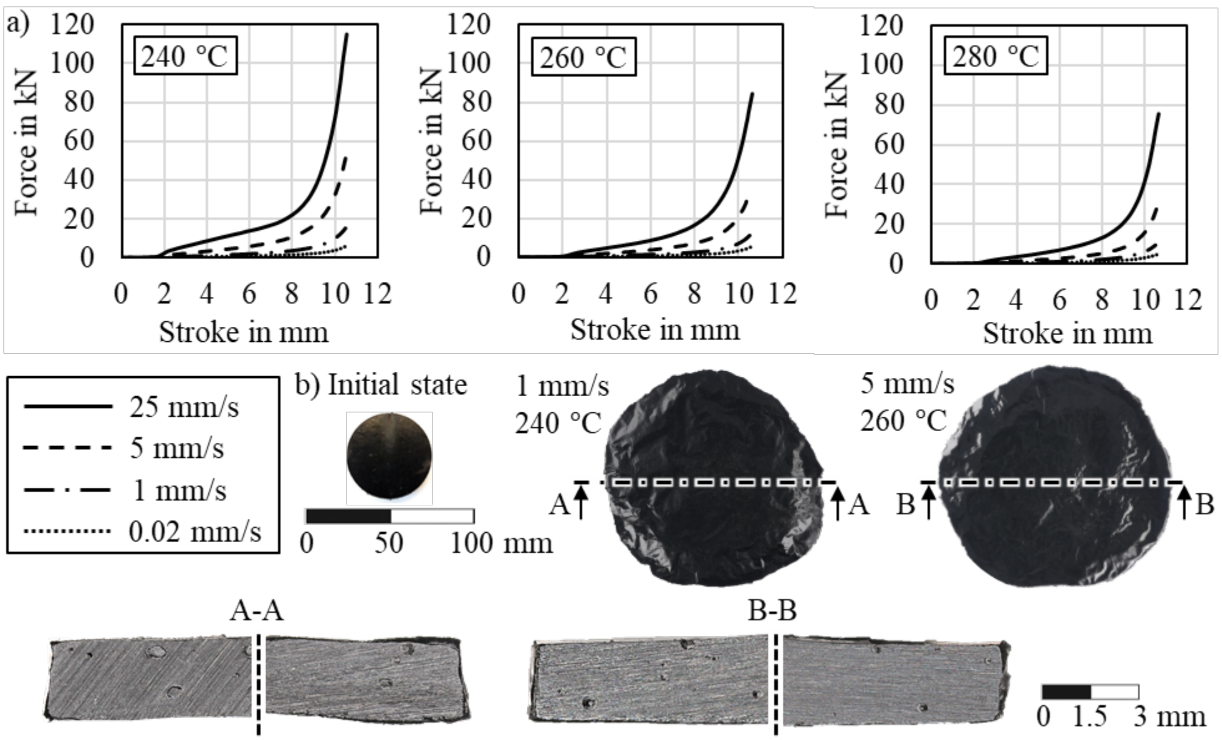

Fig. 3. Squeeze force over punch displacement for the investigated squeeze rates and temperatures a) and specimen before and after forming b).

After the compression tests, some of the specimens were cut and analysed with the digital measuring microscope VR-3200 from Keyence to obtain an indication of the shape of the flow front after forming. The sectional views in Fig. 3 b) show, that the flow fronts have nearly straight edges, which indicates a slip at the tool surface and thus a pure biaxial extension of the specimen. But it must be taken into account, that the GMT was still molten when the tool plates were opened, so that slight changes in the shape of the flow fronts may have occurred after the forming.

Parameter Fit for Shear Flow

Firstly, the compression tests are analysed assuming a pure shear flow. For this purpose, Scott's equation (Eq. 1) is used to determine the coefficients K and n for the calculation of the shear viscosity. Therefore, Eq. 1 is logarithmised to obtain Eq. 7.

$$\log(F_{Shear}) = n\ \log\left(\frac{u}{4}\right) + \log\left\{\left(\frac{2n+1}{n}\right)^n \left(\frac{2}{h}\right)^2 \frac{K\pi R^{n+3}}{(n+3)}\right\} \quad (7)$$

Subsequently, a plot of $\log(F_{Shear})$ over log(u/4) is created, which is shown in Fig. 4 a) as an example for various plate distances at 260 °C. From the diagram, the power law coefficient n can be determined as the gradient of the curves and B from the intersection point with the y-axis. It can be seen, that the coefficients vary with the plate distance h. Therefore, averaged values were calculated for each temperature, which are summarised in Table 1. In Fig. 4 b), the resulting shear viscosity are shown as a function of the shear rate for the investigated temperatures. It can be seen that the shear viscosity of the GMT decreases with increasing shear rate and temperature.

Table 1: Power law coefficient for the shear viscosity.

Temperature in °C	K in Pa s	n
240	132391.97	0.564
260	77917.00	0.512
280	48583.70	0.470

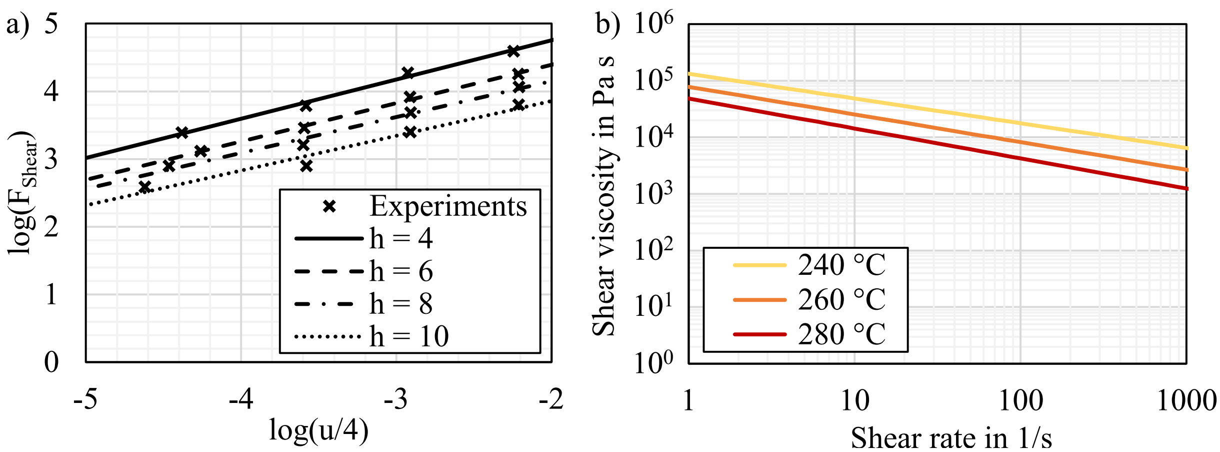

Fig. 4. Logarithmic plot for the determination of the power law coefficients a) and resulting shear viscosity over shear rate b).

Parameter Fit for Biaxial Extension

Secondly, a pure biaxial extension of the GMT was assumed and the true stress, true strain and strain rate were determined using Eq. 5. In Fig. 5, the true stress is shown as a function of true strain and strain rate for the investigated squeeze rates and temperatures. It can be seen that the stress increases with increasing strain and strain rate. In order to use the true stress-strain curves as an input for the numerical simulation, the curves must be known for a constant strain rate. This requires the modelling of the test results. For this purpose, a 4th degree polynomial approach was fitted for each temperature using the *Curve Fitting Toolbox* from *Matlab*. The fitted function with the corresponding coefficients and the coefficient of determination are given in Eq. 8 and Table 2.

$$\sigma = p_{00}+p_{10}\cdot\varepsilon+p_{01}\cdot\dot{\varepsilon}+p_{20}\cdot\varepsilon^2+p_{11}\cdot\varepsilon\cdot\dot{\varepsilon}+p_{02}\cdot\dot{\varepsilon}^2+p_{30}\cdot\varepsilon^3+p_{21}\cdot\varepsilon^2\cdot\dot{\varepsilon}+p_{12}\cdot\varepsilon\cdot\dot{\varepsilon}^2+p_{03}\cdot\dot{\varepsilon}^3+p_{40}\cdot\varepsilon^4+p_{31}\cdot\varepsilon^3\cdot\dot{\varepsilon}+p_{22}\cdot\varepsilon^2\cdot\dot{\varepsilon}^2+p_{13}\cdot\varepsilon\cdot\dot{\varepsilon}^3 \quad (8)$$

Table 2: Modell coefficients and R^2 for biaxial extension.

	p_{00}	p_{10}	p_{01}	p_{20}	p_{11}	p_{02}	p_{30}	p_{21}	p_{12}	p_{03}	p_{40}	p_{31}	p_{22}	p_{13}	R^2
240°C	-0.143	2.079	0.094	-3.050	4.839	-0.028	1.479	-3.501	-0.681	0.003	-0.195	1.613	-0.215	0.100	99.29%
260°C	1.554	1.075	1.183	-0.451	0.615	-0.333	0.150	-0.423	-0.472	0.272	0.025	0.173	-0.154	0.275	99.53%
280°C	-0.012	0.210	-0.039	1.060	1.863	0.013	-1.614	-1.327	-0.179	-0.001	0.596	0.633	-0.067	0.024	99.64%

The resulting models are shown in Fig. 5 as the coloured surfaces. These were used to calculate true stress-strain curves for constant strain rates, which were implemented in the material model for the numerical simulation. A description of the simulation model follows in the next section.

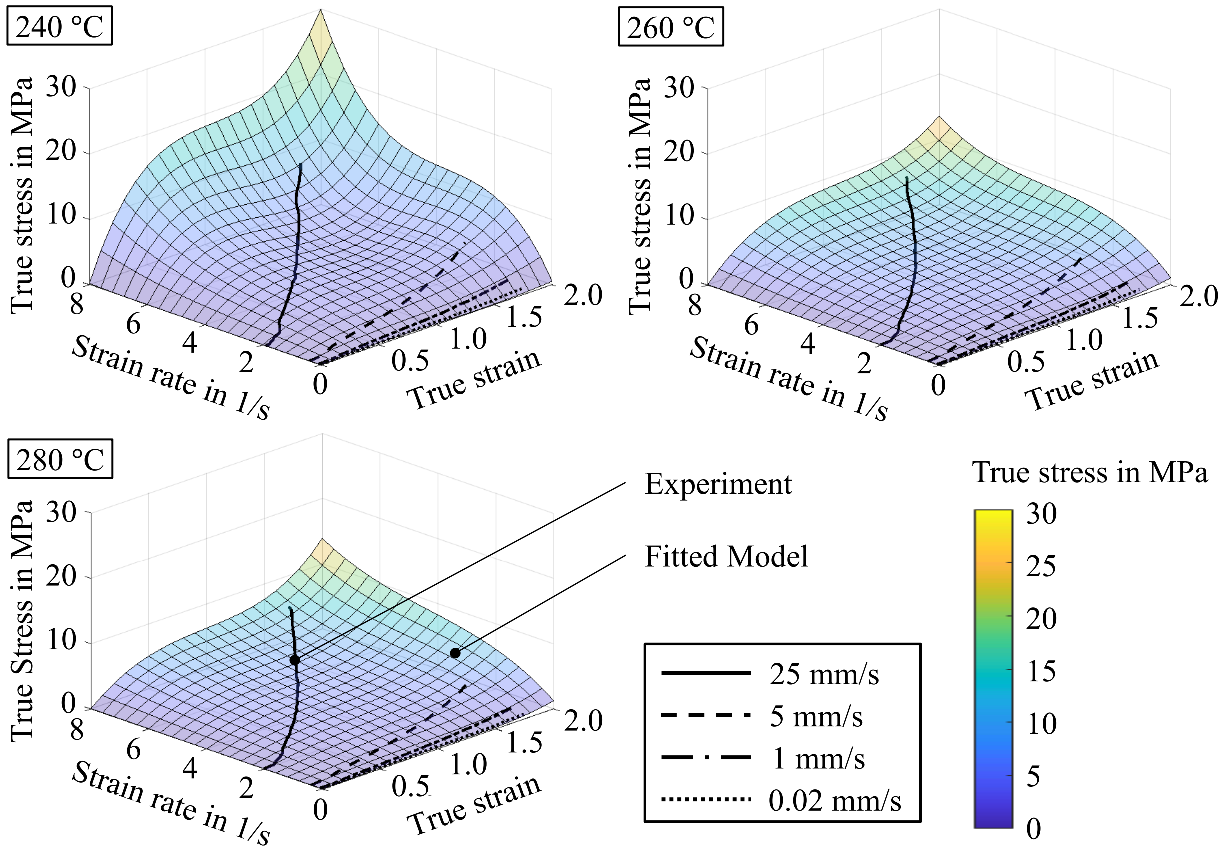

Fig. 5. True stress over true strain and strain rate for the investigated squeeze rates and temperatures.

Numerical Simulation of the Compression Tests

For the verification of the material models, the compression tests were simulated with Abaqus using the CEL method. A three-dimensional model of the compression test was created, which consists of the two tool plates, the GMT specimen and the Eulerian space in which the material can flow. To reduce the computational time, the symmetry was exploited and only one eighth of the setup was considered. The model with the corresponding components is shown in Fig. 6.

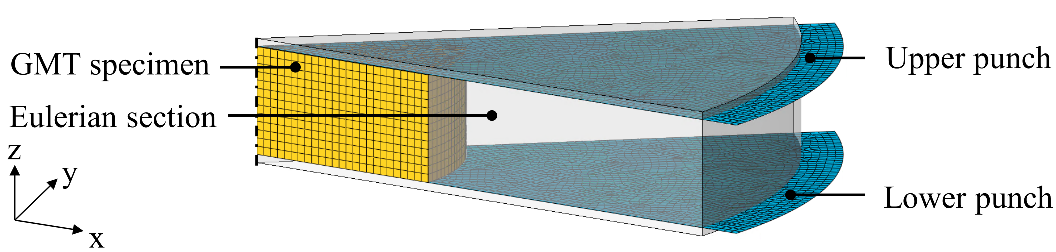

Fig. 6. Numerical model of the compression tests.

The tools were defined as ideal rigid bodies and meshed with an element edge length of 1 mm. For the Eulerian section, an element edge length of 0.9 mm was applied to ensure that at least three elements are present across the thickness at the smallest punch distance. Two material models were applied to model the flow behaviour of the GMT specimen. Firstly, the specimen was associated with a viscous material model and the shear viscosity was defined using the power law coefficients in Table 1. In the simulation with the viscous material model, a sticking contact was defined between the tool surfaces and the specimen, as the coefficients were evaluated under this assumption. In the second material model, the flow of the GMT was modelled using the true stress-strain curves shown in Fig. 5. The curves were implemented as a function of the strain rate and temperature. Here, a frictionless contact between the tools and the specimen was defined in order to ensure a complete slip of the material on the tool surface. The temperatures were defined in the simulation according to the experimental tests. For verification purposes, only the simulations with a squeeze rate of 5 and 25 mm/s were analysed, as these are in the relevant range of the later compression moulding tests in an industrial scale. Furthermore, these tests could be calculated in an acceptable amount of time due to the short test duration.

Numerical Results and Discussion

After simulation, the force-displacement curves predicted by the simulation were compared to the experimental results, which are shown in Fig. 7. If a pure shear flow is assumed, the predicted squeeze force is underestimated in all simulations up to a punch displacement of approximately 8 mm. With further increasing displacements, the predicted squeeze force increases exponentially and clearly exceeds the experimental force. This is in accordance to the findings of Kotsikos et al. [8], who calculated the squeeze force analytically. Consequently, the shearing of the material only appears to have a significant influence on the forming force when small punch distances are reached.

Under the assumption of a pure biaxial extension, the predicted squeeze force increases at low punch displacements, which results in a better agreement to the experimental curves. Overall, a higher accuracy of the predicted squeeze force is achieved up to the end of the forming process. This leads to the conclusion, that the assumption of pure biaxial flow provides a more accurate representation of the material behaviour during the compression tests. Consequently, the flow modelling using true stress-strain curves is the most promising approach in this case. Before the biaxial flow model can be applied in the simulation of the industrial process, the applicability has to be verified. If it turns out, that no complete slip occurs between the GMT and the industrial mould surface, the material model might lead to inadequate results. In this case, the compression tests should be repeated with an adhesive layer between the GMT und the tool surface, which prevents slip. This would allow the modelling of a pure shear flow or a combination of shear flow and biaxial extension.

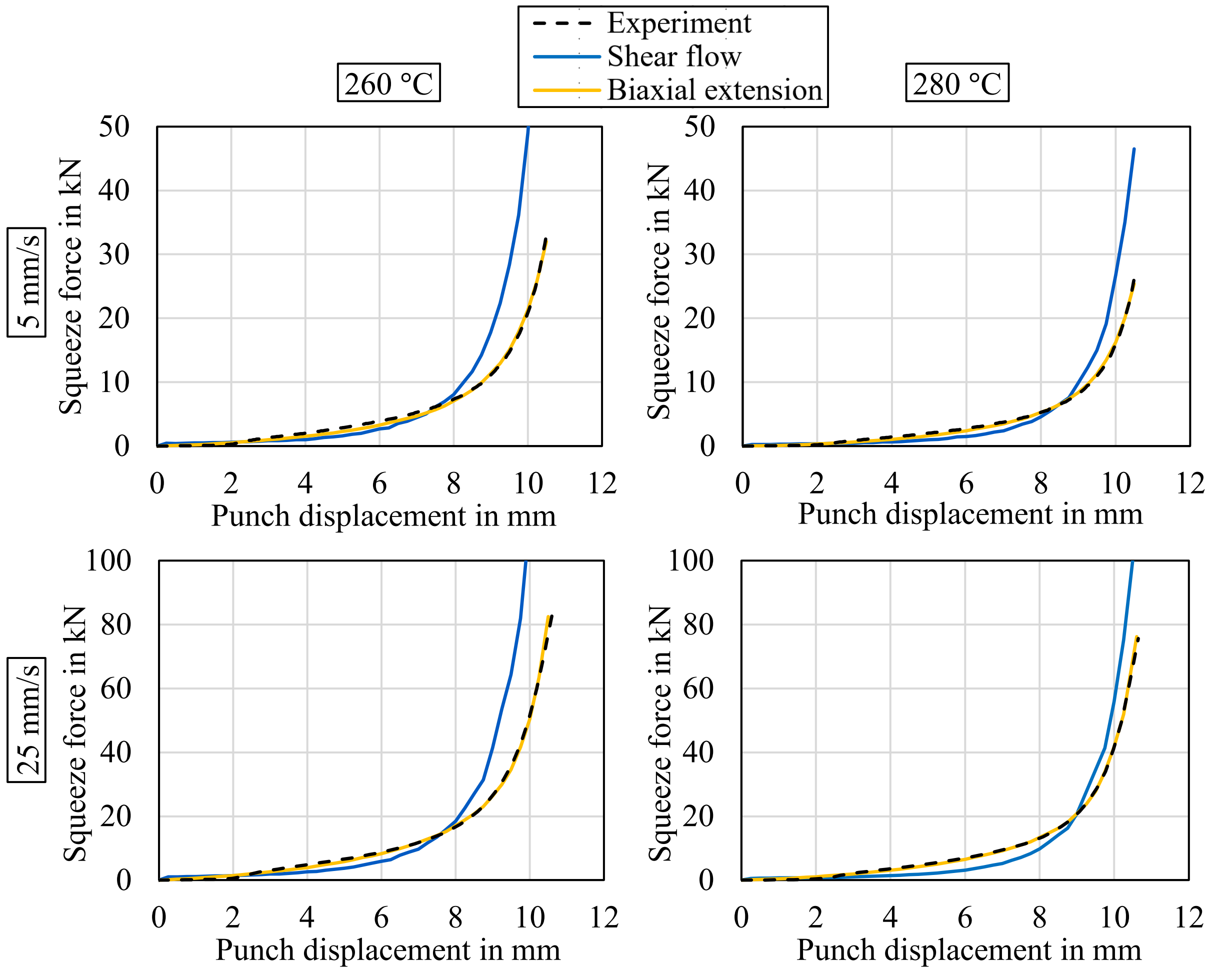

Fig. 7. Comparison of experimental and numerical squeeze forces over punch displacement.

Conclusion and Outlook

In this contribution, the rheological properties of a GMT (PA6-GF30) were determined to model the material flow in a compression moulding simulation. Isothermal compression tests were carried out in a parallel plate rheometer at varying squeeze rates and temperatures. The flow behaviour was modelled under the assumption of pure shear flow and a pure biaxial extension. It was found that the shear viscosity of the GMT decreases with increasing temperature and strain rate and the true stress increases with strain and strain rate. For a verification of the results, the compression tests were simulated in Abaqus using the CEL method. Here, the first considered assumption of a pure shear flow led to an underestimation of the squeeze force for small punch displacements and to an overestimation for high punch displacements. The second considered assumption of a pure extensional flow led to an enhanced prediction accuracy regarding the resulting squeeze force.

In future work, the material model will be used to simulate the combined forming process of the battery housing structure made of GMT and UD tapes. The UD tapes will be modelled as a composite shell lay-up. For the GMT, the CEL method will be applied, as this proved to be a suitable method for the simulation of squeeze flow. If it turns out that the biaxial extension model does not lead to reliable simulation results for the industrial process, further investigations will be carried out and the material flow will be modelled as pure shear or a combination of shear flow and biaxial extension. The objective of the industrial process simulation is to identify a suitable process window to ensure the production of high-quality components. Furthermore, the tool design can be supported and the amount of time- and cost-consuming trial-and-error tests can be reduced.

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