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The creep behavior of long fiber reinforced thermoplastics examined by microstructural simulations

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ABSTRACT

Long fiber reinforced thermoplastics (LFT) appear to be promising for the cost efficient manufacture of lightweight structures by injection or compression molding. One major concern persists in their inherent tendency to creep due to continuous sliding within the thermoplastic matrix. To enable the application of LFT components under significant static loads, a profound knowledge of the interactions between the viscoelastic matrix and the nonwoven, discontinuous fiber reinforcement is necessary. In the present work, these interactions are investigated by micromechanical finite element simulations of computer generated LFT structures. The viscoelastic properties of the neat matrix are experimentally characterized and implemented into the microstructural models by an appropriate constitutive law in the form of a four parameter Burgers model. Since a distinct degree of nonlinearity is observed, the applied model is extended to the nonlinear viscoelastic regime and found to be suitable for an accurate reproduction of the experimental data. Micromechanical creep simulations which incorporate the viscoelastic matrix behavior are then validated against creep experiments on LFT specimens of three material variants with a different fiber fraction (PPGF10, PPGF20 and PPGF30), which are loaded under two different orientations at multiple stress levels. The model predictions show a good agreement to the experimental results in particular for the lower and medium stress levels, whereas a slightly increased error can be observed for the highest stress levels. By the application of different variants of the viscoelastic matrix model it is shown that the effects of nonlinearity on the effective creep behavior of the composite are quite considerable. Finally, the evolution of stress and strain within the microstructure during the creep period is visualized by contour plots at different times. The redistribution of stress from the viscoelastic matrix to the elastic fibers can clearly be observed.

1. Introduction

Thermoplastic matrix composites like long fiber reinforced thermoplastics (LFT) gain increasing importance for the manufacturing of lightweight structures in automotive industry. They represent a good tradeoff between the mechanical performance and costs due to their suitability for mass production by technologies like injection or compression molding and their sufficiently high fiber length, resulting in a high mass related strength [1]. Furthermore, they are recyclable due to the reversibility of the solidification process of the thermoplastic matrix. However, a limitation toward the application in structural parts under significant static loads persists due to their inherent tendency to creep deformation. This results from the load driven, continuous sliding of the polymeric chains of the thermoplastic matrix. The lack of a side-chain network, as compared to thermosets, is responsible for the more pronounced creep. Whereas the viscoelastic properties of monolithic thermoplastic materials were studied to a rather high degree, only few works have been reported on fiber reinforced thermoplastics. This applies in particular to discontinuous fiber reinforced composites with a nonwoven structure. To enable their application in structural parts, a profound understanding of the microstructural interactions between the thermoplastic matrix and the fibers is of crucial importance.

A variety of viscoelastic models has been proposed in the literature. Most of them were originally developed to describe the mechanical behavior of bulk polymeric materials, but can also be applied to composites or - within a microstructural model - to

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describe the viscoelastic phase of the composite. These include the classical Maxwell and Kelvin-Voigt elements and their combination to the four parameter Burgers model. Usually, Boltzmann's superposition principle in the form of convolution integrals is applied for their mathematical description, yielding the memory function, restricted to linear viscoelastic behavior in its original form. Fundamental contributions toward the extension to nonlinear viscoelasticity by the introduction of nonlinearizing functions into the convolution integral were made by Leaderman [2]. Schapery [3] proposed a more general and thermodynamically consistent form of Leaderman's modified superposition principle, which is widely accepted today. Findley et al. [4] followed a different route by describing the time-dependent deformation by a power law, which is restricted to a single relaxation time in contrast to the Schapery model. A variety of finite element implementations of such models and their application to various loading scenarios is presented in the literature. Lai and Bakker [5] and Haj-Ali and Muliana [6] described the implementation of the Schapery model. An excellent overview and comparison of multiple models with respect to their numerical implementation was given by Woldekidan [7].

A general review of the creep behavior of polymer matrix composites (PMCs) and related experimental and modeling techniques to characterize them was published by Scott et al. [8]. Macroscopic material laws applicable to fiber reinforced plastics were presented by Lou and Schapery [9]. Greco et al. [10] studied the flexural creep of a thermoplastic composite with a woven structure and applied an analytical model to describe the experimental findings. The influence of a varying fiber volume fraction on the creep behavior of polypropylene-based composites has been examined by Houshyar et al. [11]. Howard and Hollaway [12] characterized a randomly oriented glass fiber/polyester composite and applied the Schapery model to describe the experimental data. All these approaches are restricted to the macroscopic level - the micromechanical interactions within the heterogeneous materials are not considered.

Micromechanical methods which determine the effective composite properties based on the constituent behavior are widely used to predict the elastic properties. In contrast, relatively few works extend the classical homogenization schemes to viscoelasticity. Such methods usually include a viscoelastic description of the matrix, the fibers or the interface to represent their time-dependent behavior. Analytical approaches like the Mori-Tanaka scheme [13] offer the advantage of very low computing times but require significant simplifications with respect to the geometry and the interactions of the inclusions. The analytical approaches were found to be sufficiently accurate to describe the time-independent, elastic properties of LFTs with a distinct fiber orientation and length distribution [14–16]. However, this is not necessarily the case for their viscoelastic behavior. Finite element (FE) models which explicitly depict the microstructure in the form of representative volume elements (RVEs) overcome these simplifications, but they usually require massive computational resources. Brinson and Lin [17] used the Mori-Tanaka approach to model a composite of viscoelastic phases by the application of the elastic-viscoelastic correspondence principle and compared it to FE simulations of a unidirectional composite. Brinson and Knauss [18] analyzed multiphase, viscoelastic solids by FE simulations. Matzenmiller and Gerlach [19] presented a micromechanical model of viscoelastic composites with compliant fiber-matrix bonding based on the generalized method of cells. Viscoelastic interphases were also treated by Fisher and Brinson [20]. The nonlinear viscoelastic behavior of laminated composites was examined by Haj-Ali and Muliana [21] by combining the Schapery model to describe the matrix and a four-cell micromodel to account for the unidirectional, elastic fibers.

Specific micromechanical models to describe the creep behavior of a nonwoven fiber structure with a distinct fiber orientation and length distribution are currently not available. Therefore, this work extends a microstructural model for LFTs [22,23] to include a viscoelastic description of the matrix to examine its creep behavior. For this purpose, artificial LFT structures of three materials with different fiber fractions are generated under compliance of experimentally measured microstructural data: fiber orientation distribution, fiber length distribution and fiber volume fraction (Section 3.1). The viscoelastic properties of the matrix are determined on substance specimens of the neat matrix. Based on the experimental findings, an appropriate constitutive law in the form of a four parameter Burgers model is implemented into the FE software and calibrated on the creep tests (Section 3.2). Since the experimental results reveal a strong degree of nonlinearity, the model is extended to the nonlinear viscoelastic regime. Finally, creep simulations are performed on the computer generated LFT microstructures using the viscoelastic matrix model. The predictions are compared to experimental creep tests on LFT specimens in Section 4.1. To visualize the effects of the nonlinear matrix behavior on the effective creep curves of the composite, different variants of the viscoelastic constitutive law are applied and compared. Moreover, the evolution of microscopic stress and strain is studied by contour plots of the microstructure at different times (Section 4.2). Potential applications of our model are discussed in Section 4.3.

2. LFT material and experimental methods

The investigated LFT material consists of a polypropylene matrix (*DOW® C711-70RNA*) and glass fibers (*TufRov® 4575*). Three material variants PPGF10, PPGF20 and PPGF30 were produced with different fiber weight fractions of 10, 20 and 30 wt-% (corresponding fiber volume fractions of 3.8, 8.2 and 13.2 vol-%). PPGF30 and PPGF20 are of special interest for structural application and commonly used by the automotive industry, whereas PPGF10 is rather of academic interest to provide additional data for the model validation. Plates with dimensions of $400 \times 400 \times 3$ mm^3 were manufactured by compression molding as described by Henning et al. [24]. The LFT strand as it came out of the extruder was positioned asymmetrically in the mold. The material flow during the compression of the mold generates a rather high degree of fiber orientation everywhere outside the strand inlay position. This so-called flow region of the plate was investigated in the following. For mechanical testing, tensile specimens were machined in 0° (flow direction) and 90° (transverse direction) relative to the mean orientation of the fibers. Micro computer tomographic (CT) measurements were conducted to determine the fiber orientation distribution of the material which is required for the microstructural models. The fiber length distribution was determined by analysis of incinerated specimens. More details of these procedures are given in previous contributions [22,23]. To measure the mechanical properties of the neat polypropylene, matrix substance specimens were manufactured by injection compression molding. To ensure the mechanical compatibility between the matrix specimens and the matrix within the LFT samples, the same coupling agents and stabilizers as used in the LFT process were added for the matrix substance specimens.

Creep tests on matrix and LFT specimens (geometry according to DIN EN ISO 3167 with a reduced section of $70 \times 10 \times 3$ mm^3) were performed at standard climate (23 °C temperature, 50% relative humidity). At the beginning of the load period of $6 \cdot 10^5$ s, a load defined by the product of specimen cross section and desired nominal stress level was applied in the form of a lead weight. For the lower stress levels up to 5 MPa, the load was directly attached to the specimen clamps, whereas a lever mechanism with a transmission ratio of 10 was used for the higher stress levels. The strain