A micromechanical approach to time-dependent failure in composite systems

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Abstract

The time-dependent failure behaviour of off-axis loaded composites is investigated, assuming that fracture is matrix dominated. Since the stress and strain state of the matrix in composite structures is complex, the yield and fracture behaviour of a neat epoxy system is investigated under various multi-axial loading conditions. A good description of the multi-axial yielding behaviour of the matrix material is obtained with the 3-dimensional pressure modified Eyring equation. The parameters of this 3-dimensional yield expression are implemented into a constitutive model, which has been shown to describe the deformation behaviour of polymers under complex loadings correctly. By means of a micromechanical approach, the matrix dominated transverse strength of a unidirectional composite material was investigated. Numerical simulations show that a failure criterion based on maximum strain provides a good description for the rate dependent transverse strength of unidirectional glass/epoxy composites. Furthermore, such a strain criterion is also able to describe the durability (creep) of off-axis loaded unidirectional composites.

1. Introduction

Models for the prediction of strength of composite materials are generally directed to short-term failure using laminate analysis based on classical mechanics, coupled with a common failure theory such as maximum stress or maximum strain concepts (Tsai & Hahn, 1980). The input data for these models is derived from standard tests on unidirectional laminates, normally in accordance with the American Society for Testing and Materials (ASTM) standards, and are typically low strain rate or quasi-static experiments. Subsequently, this data is often used by designers to analyze structures which are subjected to long-term static (creep), dynamic (fatigue) or shock (impact) loadings, which all differ strongly from the standard test conditions.

In the field of polymer physics it is well established that the deformation as well as the failure behaviour of polymers is strongly influenced by the time-scale of the experiment (Brown 1986, Ward 1983). It is therefore eminent that composites based on these materials will also display a large influence of the time-scale, especially in off-axis loading situations where the properties are strongly governed by the viscoelastic polymer matrix. With respect to durability this implies that the mere fact that a composite material is loaded well below the critical stress, as determined in a short-term test, will not ensure that the material will sustain this load for an infinite period of time.

One of the most important factors determining the failure behaviour of glassy polymers is yielding and the ability or inability to plastically deform. Unlike metals, the development of plastic deformation will occur in polymers at any stress level. Under the influence of an applied stress the development of plastic deformation will start immediately upon loading, and will proceed at a constant rate that depends strongly on the stress level. In a tensile test, the rate of plastic deformation
will gradually increase with increasing stress until it is in balance with the externally applied strain rate and a constant level of stress is reached: the yield stress. With decreasing strain rate, or, equivalently, increasing temperature, this yield stress will decrease (Brown 1986, Ward 1983).

In the present research the transversal failure behaviour of a glass-fibre/epoxy composite is studied in constant strain rate and creep experiments in three-point bending. The experimental results are subsequently analysed employing micromechanics. To facilitate this analysis the deformation behaviour of the epoxy matrix is investigated in detail. Since previous studies already showed the importance of a multi-axial stress state for the initiation of transversal failure (Asp & Berglund 1995), the yield behaviour of the epoxy matrix is characterised in various multiaxial loading conditions. The experimental data is analysed using a pressure-modified Eyring equation (Ward 1971, Duckett et al. 1978).

With recent developments in three-dimensional constitutive modelling of large strain plasticity in amorphous polymers (Boyce et al. 1988 & 1994, Wu & van der Giessen 1993, Tervoort et al. 1996 & 1998), the numerical simulation of the behaviour of the polymer matrix under complex loading conditions is now well established. In the present work a previously developed constitutive model, the so-called compressible Leonov model (Tervoort et al. 1996 & 1998), was employed in the micromechanical finite element simulations. In combination with a maximum strain criterion for the epoxy matrix, it is attempted to describe the rate dependent strength and creep life-time of a 10° and 90° off-axis loaded unidirectional glass/epoxy composite.

2. Experimental

2.1 Materials

For the matrix material a rather brittle epoxy system of Ciba Geigy (Araldite) was used. This epoxy system is based on diglycidyl ether of bisphenol-A (LY556) with an anhydride curing agent (HY917) and an accelerator (DY070) in the weight ratio of 100:90:1. In this study E-glass fibres (Silenka 0.84-M28) were used as reinforcement material.

Samples for experiments on the neat epoxy where milled from plate material. For the fabrication of these plates, the constituents of the epoxy system were carefully mixed. The matrix material was subsequently poured into a preheated aluminium casting-mould and was cured for 4 hours at 80°C and 8 hours at 140°C. Plates (240x180 mm) were made with a thickness of 2, 4 and 6 mm.

Unidirectional composites were manufactured by filament winding. Fibre strands were impregnated in a heated epoxy bath and wound uniformly on a framework. The impregnated fibres were subsequently placed in a mould and cured in a hot press for 4 hours at 80°C and 8 hours at 140°C. The resulting composite plates had a thickness of 1.25 mm and a fibre-volume fraction of approximately 50%.

From these unidirectional laminates, test specimens were cut with a fibre direction of 10° and 90°. In the case of 10° tensile specimens aluminium end-tabs were adhesively bonded to the specimens. The 10° off-axis tensile specimen had a length between the tabs of 200 mm and a width of 12.5 mm. Transverse (90°) specimens where tested in three-point bending. These three-point bending specimens where cut at a length of 50 mm and a width of 25 mm, as suggested by ASTM D790M-92. The edges of all samples were manually finished using SiC polishing paper (up to 4000 FEPA Standard).
2.2 Testing

The experimental part of this study consisted of mechanical test on pure matrix material and on unidirectional composites. In order to characterize the yield behavior of the matrix under complex stress fields, various tests are needed. In this study the material was studied at strain rates varying over several decades in five different loading geometries: uniaxial extension, uniaxial compression, planar extension, planar compression and simple shear. All experiments were performed at room temperature.

Uniaxial tensile tests were performed on 2 mm thick dog-bone specimens, according to ASTM D638-91. Uniaxial compression samples were end-loaded and measured 10x6x6 mm. The planar extension samples were dog-bone shaped, with a testing section of 50 mm wide, 10 mm long and 2 mm thick. Due to the high width-to-length ratio the contraction of the material is constrained in the width of the sample, which creates a plane strain condition during the straining of the sample (Whitney & Andrews 1967). Planar compression tests were performed on dog-bone shaped specimens with a test section measuring 85x20x4 mm. In the test section the specimens were supported by steel plates to create a plane strain condition. Simple shear tests, according to ASTM D4255-83, were performed on samples with a thickness of 2 mm and a length of 100 mm. The distance between the grips was 10 mm, resulting in an aspect ratio of 10.

In the case of the $10^3$ off-axis samples uniaxial tensile tests and creep tests where performed on a Frank tensile machine (type 81565) at room temperature in normal environmental conditions. The uniaxial tensile tests where performed at strain rates varying from $10^{-5}$ to $10^2$ s$^{-1}$. To determine the $90^\circ$ off-axis properties of the unidirectional glass fibre reinforced composites, three-point bending tests were performed in accordance with ASTM D790M-92. The strain rate in the outer layers of the samples in the three-point bending test varied from $1.7 \times 10^{-6}$ to $1.7 \times 10^{-3}$ s$^{-1}$. A support length of 32 mm and thickness of 1.25 mm resulted in a span-to-depth ratio of 25. The strain rate and creep experiments were both performed on a Frank 81565 tensile tester.

3. Constitutive modelling

3.1 The compressible Leonov model

In previous work an elasto-viscoplastic constitutive equation for polymer glasses was introduced, the so-called compressible Leonov model (Tervoort et al. 1996 & 1998). For the present work the model is extended include both the influence of pressure on the yield stress and the strain hardening effect. In that case the Cauchy stress tensor $\sigma$ is considered as a parallel assemblage of two distinguishable parts: The driving stress tensor $s$ and the hardening stress tensor $r$ respectively:

$$\sigma = s + r$$

(1)

The expression for $s$ is obtained from the compressible Leonov model (Tervoort et al., 98), in that case $s$ is decomposed in a hydrostatitc and a deviatoric part, and related to the deformations according to:

$$s = K(J - 1)I + G\tilde{B}^d$$

(2)
where $K$ is the bulk modulus and $G$ is the shear modulus. The relative volume change $J$ and the isochoric elastic left Cauchy Green deformation tensor $\tilde{B}_e$ are implicitly given by (Tervoort et al. 1998):

$$J = \text{tr}(D)$$

(3)

$$\tilde{B}_e=\left(D^d - D_p^d\right) \cdot \tilde{B}_e + \tilde{B}_e \cdot \left(D^d - D_p^d\right)$$

(4)

The left hand side of this equation represents the (objective) Jaumann derivative of the isochoric elastic left Cauchy Green tensor. The tensor $D$ denotes the rate of deformation tensor, $D_p$ the plastic rate of deformation tensor. To complete the constitutive description the plastic deformation rate is expressed in the Cauchy stress tensor by a generalized non-Newtonian flow rule:

$$D_p = \frac{s^d}{2\eta(\tau_{eq}, p)}$$

(5)

where $\tau_{eq}$ and $p$ are parameters to be defined in the following. The viscosity $\eta$ is derived assuming pressure modified Eyring flow (Ward 1971, Duckett et al. 1978):

$$\dot{\gamma}_{eq} = \frac{1}{A} \sinh \left[ \frac{\tau_{eq}}{\tau_0} \right] \exp \left[ -\frac{\mu p}{\tau_0} \right]$$

(6)

where $\dot{\gamma}_{eq}$ is the equivalent shear rate, $\tau_{eq}$ is the equivalent shear stress and $p$ is the hydrostatic pressure, according to:

$$\dot{\gamma}_{eq} = \sqrt{\text{tr}(D_p \cdot D_p)}$$

(7)

$$\tau_{eq} = \frac{1}{2} \text{tr}(s^d \cdot s^d)$$

(8)

$$p = -\frac{1}{3} \text{tr}(s)$$

(9)

In this case, the viscosity function $\eta$ can be expressed as:

$$\eta(\tau_{eq}, p) = A \tau_0 \exp \left( \frac{\mu p}{\tau_0} \right) \frac{\tau_{eq}/\tau_0}{\sinh(\tau_{eq}/\tau_0)}$$

(10)

where $A$ is a time constant, $\tau_0$ a characteristic stress and $\mu$ is a pressure coefficient. The hardening behavior of the material is described by the hardening stress tensor $r$. 


In the present work the hardening stress was modelled as a neo-Hookean spring. In the assumption that the network is incompressible, the neo-Hookean relation ship for the hardening stress tensor \( r \) can be written as:

\[
 r = G_R \tilde{B}^d
\]  

(11)

with \( G_R \) the strain hardening modulus (assumed temperature independent).

### 3.2 Material characterisation

**Elastic parameters**

The elastic modulus \( E \) and the Poisson’s ratio \( \nu \) of the matrix material were 3200 MPa and 0.37 respectively, as determined in a tensile experiment. The elastic properties of the glass fiber were assumed \( E=70 \) GPa and \( \nu=0.22 \).

**Yield Parameters**

The results of the uniaxial extension, planar extension, uniaxial compression, planar compression and simple shear test, all performed at a temperature of 22°C, are presented in Figure 1. In this Figure the experimental yield stress and strain rate were converted into the equivalent shear stress and strain rate respectively. Figure 1 clearly demonstrates the pressure dependence of the material since the results of the different loading geometries are not the same. This is the influence of the parameter \( \mu \), which increases the equivalent shear stress at the yield point with increasing hydrostatic pressure. In the absence of this pressure dependence all loading geometries would yield the same plot of \( \tau_{eq} \) versus \( \dot{\gamma}_{eq} \).

The solid lines in Figure 1 are predictions employing equation (6) with \( A= 8.2 \times 10^{20} \) s, \( \tau_0 = 1.426 \) MPa and \( \mu = 0.126 \). It is clearly shown that the model predictions with the pressure-modified Eyring equation are in good agreement with the experimental data.

**Hardening Modulus**

The hardening parameter \( G_R \) is, in the present study, determined in a uniaxial compression test. Figure 2 shows the result of a uniaxial compression experiment at a strain rate of \( 10^{-4} \) s\(^{-1} \). In this figure the axial true stress \( \sigma \) is plotted as a function of the strain measure \( \lambda^2-\lambda^{-4} \). This strain measure is, in a uniaxial compression (or tensile) test, the component of the deviatoric isochoric left Cauchy green deformation tensor \( \tilde{B}^d \) in load direction. According to the Neo-Hookean approach, the compression curve should display a constant slope after yield.

As shown in Figure 3, this assumption is valid up to compressive strains of approximately 15%. At higher strain levels the slope is increasing, indicating a possible influence of the limited extensibility of the epoxy network. In the present work, the Neo-Hookean approach was chosen, since it was readily available in the finite element code. From Figure 3, the value of the strain hardening modulus \( G_R \) was determined to be 10.5 MPa.
Figure 1: The equivalent shear stress versus equivalent shear rate for the various test conditions. The solid lines are predictions according to equation (6) with the parameters listed in the text.

Figure 2: True stress versus $\lambda^2 - \lambda^{-1}$ for a compression test on the neat epoxy matrix at $10^{-4}$ s$^{-1}$. The dashed line indicates the fit according to the Neo-Hookean strain hardening model.
4. Micromechanical analysis of composite materials

Micromechanics is an effective tool that can be employed to estimate the mechanical or physical properties of a composite material on the basis of the properties of its constituents and their microstructural ordering. To analyse a microstructure by a micromechanical finite element simulation, there are basically two requirements: 1) the choice of an appropriate constitutive law, in our case the compressible Leonov model discussed in the previous section, and 2) the definition of a representative volume element that accounts for the microstructural arrangement of the components. In our case, where its attempted to analyse (time-dependent) failure phenomena, an additional failure criterion needs to be chosen and quantified.

4.1 Finite element model

In the case of off-axis loaded unidirectional composites, the microstructure is characterized by a random arrangement of fibres in a polymer matrix. Since a detailed geometrical model of this microstructure would lead to excessive computing time, especially in the case of time-dependent behaviour, the topography is usually simplified by assuming a more regular stacking of the fibres. A successful simplification is to assume that the fibres are stacked regularly in a hexagonal array, from which the repeating unit is shown in Figure 3a. Due to symmetry only a quarter of this repeating unit needs to be modelled, which leads to a decrease in elements and thus simulation time. To be able to perform simulations under various loading angles between fibre and load, a 3D-mesh has to be used. This 3D-mesh represents a fibre-volume fraction of 50% and consists of a single layer of 507 3D bilinear 20 noded reduced integrated elements. The mesh is shown in Figure 3b, as seen from a direction parallel to the fibers.

![Figure 3: (a) Representation of the hexagonal stacking pattern. (b) Transeversely loaded 3D RVE-mesh. Other off-axis angles are obtained by rotating this mesh around the z-axis.](image-url)
To facilitate the finite element analysis of the off-axis loading at constant strain rate as well as at constant load, we used a micro-macro coupling procedure developed by Smit et al. (1998). In this multi-level finite element method the analysis is performed on two levels: a macroscopic and a microscopic level. The first level, the macroscopic level, is the level at which the loads are applied. The second level is the micro level, identified by a representative volume element (RVE), which is in fact a finite element model of the microstructure (in our case the mesh presented in Figure 3b). To enforce periodicity of the RVE during and after deformation, two opposite sides must deform in exactly the same way and stress continuity should be guaranteed across the boundaries. This is achieved by application of periodic boundary. The local macroscopic stress is obtained by applying the local macroscopic deformation (represented by the deformation tensor) on the RVE and averaging the resulting RVE stress field (determined by a finite element analysis on the RVE) (Smit et al. 1998).

The macroscopic mesh used in the present study is shown in Figure 4. As the macroscopic stress and strain fields are both homogeneous, the macroscopic structure is discretized by a single element. A linear 2 dimensional plane stress element with four integration points is used with boundary conditions as indicated in Figure 4. As the deformation is homogeneous over the element, all integration points will show the same state and history. Therefore only one macroscopic integration point is linked to the RVE and the result of the microscopic analysis is returned to all four integration points. This method is used to simulate both the constant strain rate as well as the creep loading situations. The simulation of the different off-axis orientations 10° and 90° is obtained by rotation of the RVE with respect to the macroscopic mesh.

4.2 Failure criterion: a hybrid experimental/numerical approach

With the geometrical assumptions and the choice of the constitutive behaviour, we are now able to numerically simulate the mechanical response of unidirectional composites in off-axis loading. However, since there are no restrictions to the deformation within the constitutive model, an additional failure criterion for the matrix material has to be defined. For the prediction of failure at a certain strain rate or under the influence of a statically applied global stress (creep) this criterion is employed to detect local failure in the representative volume element during a simulated loading sequence.
In this study a critical strain criterion was chosen, more specifically a critical value for the equivalent shear strain, being a second invariant of the total strain tensor:

$$\gamma_{eq} = \frac{2}{3} \sqrt{\left( (\varepsilon_1 - \varepsilon_2)^2 + (\varepsilon_2 - \varepsilon_3)^2 + (\varepsilon_3 - \varepsilon_1)^2 \right)}$$

(12)

In traditional micromechanics the critical value of the failure criterion is based on experiments on the neat matrix material. However, it can easily be anticipated that this method will not lead to the quantitative prediction of time-dependent off-axis failure aimed for in this study. The reason for this anticipated discrepancy is that the present micromechanical analysis is based on a hexagonal stacking pattern instead of the random stacking pattern that is characteristic for a real composite. In the actual case local variations in fibre-volume fraction, flaws, and voids will strongly influence the local stress situation in the matrix, and global fracture will be initiated from these imperfections. With respect to the aim of this study being lifetime prediction of composite materials, this is especially the case, since relative small changes in the (local) stress level will lead to large changes in local plastic deformation rates and thus strong variations in lifetime predictions. In the micromechanical simulations, based on a regular stacking pattern, the influence of imperfections is omitted and consequently the strength and lifetime of the composite will be overestimated when based on the true matrix strength.

As an alternative route, we introduce a hybrid experimental/numerical technique. In this approach the critical value of the failure criterion is ‘calibrated’ on a reference experiment. In this technique a reference experiment - in our case a uniaxial tensile test on a $10^\circ$ axis sample and a strain rate of $10^{-4}$ s$^{-1}$ - is chosen and the mean value of the experimentally determined tensile strength is used for the calibration. Subsequently a micromechanical simulation of an RVE with this off-axis angle is performed, applying the reference strain rate, until the averaged Cauchy stress in the direction of the load in the simulation equals the experimentally obtained tensile strength. Now the maximum equivalent strain $\gamma_{eq}$ of the simulated structure is determined and its value is assumed to be critical for matrix failure. During other simulations this maximum equivalent strain value is used as a failure criterion by assuming that crack initiation occurs when the maximum equivalent strain reaches this critical value.

5. Model validation

5.1 Rate dependent off-axis strength

The $10^\circ$ off-axis composites were tested in uniaxial tension and the $90^\circ$ transverse specimens where tested in three point bending at constant strain rate. All tests were performed at room temperature and showed perfect linear elastic behaviour up to failure. The results of maximum stress versus strain rate of the composites are presented in Figure 5. The error bars indicate the standard deviation of the experimental outcome.

As mentioned above, the mean $10^\circ$ off-axis strength measured at a strain rate of $10^{-4}$ s$^{-1}$ was used as a reference to determine the critical value of the equivalent shear strain. For this purpose a multi-level simulation of this experiment was performed, and stopped at a macroscopic stress level of 290 MPa, being the mean value of the experimentally determined strength. The distribution of the equivalent shear strain, at this macroscopic stress level of 290 MPa, is shown in Figure 6. The maximum value of the equivalent shear strain is 9 % and is located in a small shear band close to the fibre-matrix interface, as indicated in Figure 6. The maximum equivalent strain is localized at the pole.
Figure 5: Off-axis strength versus strain rate. The experimental results of the composite laminates (symbols) are compared to numerical simulations using the equivalent strain criterion (lines).

Figure 6: Contour plot of equivalent strain, showing the strain fields of the RVE for the reference simulation at 10° off-axis loading and a strain rate of $10^{-4}$ s$^{-1}$, at the moment that the experimentally determined macroscopic strength of 295 MPa has been reached.
With this critical value obtained, it is now possible to predict the off-axis strength at other strain rates and other off-axis angles. The predicted strain rate dependence of the $10^0$ off-axis strength is represented by the solid line in Figure 5. This line is a least square fit through the three results obtained from simulations at strain rates of $10^{-3}, 10^{-4}$ and $10^{-5}$ s$^{-1}$. As mentioned earlier the $10^0$ off-axis strength at $10^{-4}$ s$^{-1}$ is chosen as a reference and hence the experimental result coincides with the result of the simulation for this strain rate and fibre orientation. Micromechanical simulations of tensile experiments at different strain rates loads were performed, and failure was predicted using the equivalent shear strain criterion. From Figure 5 it is clear that the numerical prediction of strength is within the standard deviation for all experimental strain rates.

5.2 Creep lifetime

In this case the unidirectional composites were loaded with a constant force in either uniaxial tension or three-point bending. In all creep experiments the time needed to apply the constant engineering stress was negligible in comparison to the obtained creep time to failure. The applied stress was varied from 50 to 85 MPa for the transverse three-point bending tests and from 225 to 275 MPa for the $10^0$ off-axis tensile test. The results of the experiments are presented in Figure 7.

Similar to the constant strain rate tests, the micromechanical model, in combination with the critical equivalent shear strain of 9%, was employed to predict the creep time to failure for the different applied engineering stresses and fibre orientations. Micromechanical simulations of creep experiments at different loads were performed, and after each time-increment the maximum value of the equivalent shear strain was compared to the critical value. For the $10^0$ off-axis orientation the prediction of creep time to failure is in good agreement with the experimental data. A decrease in creep load of 7.5 MPa results in a 10 times longer time to failure. For the $90^0$ off-axis orientation the prediction of time to failure fits the experimental results for creep loads between 60 and 70 MPa. For higher creep loads the simulation tends to predict a time to failure that is on the lower side of the experimental data. The simulations show a 10 times longer life time when the load is decreased with 4.8 MPa.

![Figure 7: The applied stress versus time to break. The results of creep on off-axis loaded unidirectional composites (symbols), compared to simulations (solid line) using the equivalent strain criterion.](image-url)
In Figure 8 an example of the deformed microstructure is shown for the $10^\circ$ off-axis simulation with a creep load of 235 MPa. The deformation is multiplied with a factor 5, and shows the heterogeneous mechanical behavior of the structure. The macroscopic strain $\varepsilon = 0.9\%$ when the critical microscopic strain $\gamma_{eq, local} = 9\%$ is reached.

6. CONCLUSION

In this study it is demonstrated that the modified Eyring equation can be satisfactory used for the description of the yield behaviour of an epoxy system under multi-axial loading conditions. The various multi-axial experiments also showed that the epoxy system clearly exhibits a pressure-dependent yield behaviour.

By introduction of these parameters into the compressible Leonov model, numerical (FEM) simulations of the mechanical behaviour of the epoxy system could be performed. In combination with a representative volume element, based on a hexagonal stacking of the fibres, a micromechanical model was obtained. This model was subsequently employed to analyse time-dependent failure of unidirectional composites in transverse loading.

The critical value of the failure criterion, based on a local maximum of the equivalent shear strain, was determined by micromechanical evaluation of a reference experiment. The critical value of the equivalent shear strain obtained by this hybrid experimental/numerical technique was subsequently used for micromechanical simulations of rate dependent transverse strength and creep time to failure. In both cases the numerical simulations were in good agreement with experimental observations.
7. References


